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ASSESSMENT OF CARBON—14 CONTROL TECHNOLOGY AND COSTS FOR THE LWR FUEL CYCLE

U.S. ENVIRONMENTAL PROTECTION AGENCY

Office of Radiation Programs

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The report is an effort to incorporate present knowledge of carbon-14 behavior in Light Water Reactors and Fuel Reprocessing Plants into Designs compatible with present technology. The impact of radioactive effluents are considered according to the traditional measure of maximum radiation dose to individuals, summation of individual annual doses to obtain a total population dose, and the environmental dose commitment. The sources of carbon-14 in LWR's and fuel reprocessing facilities are identified. Systems for the removal of carbon-14 in existing and future plants are addressed from both a technological and economic standpoint. Existing technology indicates that caustic scrubbing is the most cost-effective alternative for concurrently removing C-14 from waste-gas streams and leaving it in a form compatible with permanent disposal conditions.

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ASSESSMENT OF CARBON-14 CONTROL TECHNOLOGY AND COSTS FOR THE LWR FUEL CYCLE

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FOREWORD

The Office of Radiation Programs carries out a national program designed to evaluate the exposure of man to ionizing and nonionizing radiation, and to promote the development of controls necessary to protect the public health and safety and assure environmental quality.

Office of Radiation Programs technical reports allow comprehensive and rapid publishing of the results of intramural and contract projects. The reports are distributed to groups who have known interests in this type of information such as the Nuclear Regulatory Commission, the Department of Energy, and State radiation control agencies. These reports are also provided to the National Technical Information Service in order that they may be readily available to the scientific community and to the public.

Comments on this analysis as well as any new information would be welcomed; they may be sent to the Director, Criteria and Standards Division (AW-460), Office of Radiation Programs, U.S. Environmental Protection Agency, Washington, D.C. 20460.

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CHAPTER 1. INTRODUCTION

Under the President's Reorganization Plan No. 3 of 1970, the Environmental Protection Agency was vested with responsibility for establishing environmental radiation standards. In so doing, the Agency must address public health and environmental concerns associated with the nuclear fuel cycle as a whole. In order to establish the standards on a sound basis, the following assessments must be made: comprehensive determination of the releases of radioactive materials during routine operation (planned releases) from all facilities associated with nuclear power generation, potential effects on the public and environment, minimization of these effects through the issuance of standards, and the costs and tradeoffs involved.

The impact of radioactive effluents have been considered from three points of view:

- the traditional measure of maximum radiation dose to individuals
- summation of individual annual doses to obtain a total population dose (this is equivalent to summing individual potential health effects under the assumption of a zero threshold linear relationship between dose and potential health effect)
- "the environmental dose commitment"

The latter point of view came about as a result of the observation that certain nuclides have very long half lives and so may deliver doses to populations for periods ranging from decades to millenia as they migrate through the biosphere. (1) The potential public health hazard of carbon-14 was highlighted when accounts of environmental dose commitment were made.

In response to the need for information relevant to quantitation and mitigation of the hazard, preliminary and in-depth studies have been made on the extent of carbon-14 production, environmental release and dose commitment (2, 3). Remaining was a need to review current waste processing in the nuclear fuel cycle, propose treatment options and estimate costs for available designs. The present report is an effort to incorporate current knowledge of carbon-14 behavior in light water reactors and fuel reprocessing plants into designs compatible with present technology. This information is reflected in the economic analyses presented in the final part of this study.

In summary, the purpose of this technical assessment is:

- to provide a data base for analysis for control of carbon-14 from LWR fuel cycle facilities
- to provide the basic information needed to perform a cost-effectiveness analysis for control of carbon-14 in the LWR fuel cycle
- to be an aid to the Environmental Protection Agency in establishing effluent discharge limits for carbon-14 and in reviewing environmental impact statements

Available literature on quantities and pathways of carbon-14 in fuel cycle facilities has been reviewed to analyze the behavior of this nuclide. In general, it was shown that very little carbon-14 probably remained in liquid effluent systems, so this report is much more specific to gaseous behavior and treatment; liquid cases are considered if they represent an appreciable contribution to environmental contamination. Treatment principles and devices have been reviewed and reasonable alternatives chosen. These choices have been analyzed according to conclusions regarding behavior of the nuclide and any stable carrier substances. Designs are proposed for treatment systems and waste management options considered. Finally, an economic analysis is provided and system reliability commented upon. Judgements regarding integration of various treatment and waste management alternatives are provided throughout the report. The latter task is subject to great variations as conditions of licensing, power generation and technology change, so these judgements are not to be construed as final.

Chapter 1 References

1. *Environmental Radiation Dose Commitment: An Application to the Nuclear Power Industry*, EPA-520/4-73-002, U.S. Environmental Protection Agency, February 1974.

2. Magno, P.J., C.B. Nelson and W.H. Ellett. "A consideration of the significance of carbon-14 discharges from the nuclear power industry". *Proceedings of the Thirteenth AEC Air Cleaning Conference, 1975*.

3. Fowler, T.W., R.L. Clark. J.M. Gruhlke and J.L. Russell. *Public Health Considerations of Carbon-14 Discharges from the Light-Water-Cooled Nuclear Power Reactor Industry*, Technology Assessment Division, Office of Radiation Programs, United States Environmental Protection Agency, ORP/TAD-76-3, July, 1976.

CHAPTER 2. PROJECTED CARBON-14 CONCENTRATIONS AND BEHAVIOR IN THE VARIOUS LWR EFFLUENT TREATMENT SYSTEMS

An analysis has been undertaken to estimate carbon-14 concentrations and chemical forms expected in the boiling and pressurized water varieties of light water reactor. This information will be used in design of effluent control systems. Knowledge of distribution and quantity of releases from the several pathways which carry carbon-14 to the environment allows the designer to choose cost-effective treatment principles and devices.

Deferring to design criteria established prior to and independently of this discussion, all carbon-14 produced in the reactor and released at the site will be assumed to exist in only physical and chemically bound state at the time it is removed. The chemical form of C-14 in release pathways varies, but minimum cost and technical ease dictate that most bound carbon, regardless of the state or states it passes through prior to treatment, shall be oxidized to CO_2 for extraction. Therefore, the prior forms of bound carbon-14 are important only as they would yield to ready or awkward conversion to CO_2. Conceivable forms of bound carbon in the coolant of a light water reactor are CO (gas), CO_2 (gas), and light hydrocarbons such as methane, ethane, propane and possibly butane. Kunz(4,7) reports the former two bound forms predominate in BWR's, while the latter are most significant in the PWR. The hydrocarbons are easily converted to an oxidized form.

The magnitudes of radioactive carbon source terms and their distribution among release pathways are the primary objects of this analysis. Literature pertaining to C-14 production in and release from reactors and spent fuel reprocessing plants, such as the paper by Magno, et al. of the Environmental Protection Agency,(9) was reviewed. It acknowledges that specific data and analyses are scarce. Therefore, sources for this discussion are quite recent analyses offered by researchers and regulatory agencies.(1-8) Some of the data were taken from operating experience and experiments at reactor facilities, though many of the results have been deduced from design conditions and indirect measurements. In addition, a standardized reactor size was used in this analysis, so all final values must be considered estimates of the radioactive carbon source terms. These estimates have been found very adequate as design data used in later stages of this study where approximate ranges and costs of control are established.

2.1 Sources of carbon-14 in light water reactors

This section discusses radioactive carbon sources common to both types of light water reactor. A common element of the two is the fuel rod, here the processes of nuclear fission and decay initiate production of carbon-14 in excess of its natural concentration. Direct observation of C-14 production rates in the fuel and fuel rod gap is experimentally impracticable, so reasonably accurate calculations are used to estimate the source terms. This analysis of the source terms relies upon, and follows quite closely, the work of Fowler, et al. provided in an Environmental Protection Agency (EPA) technical note.(8) Every effort has been made to use the results of the EPA study so this Technical Assessment might play a sequential role in any discussion of the extent, ramifications and mitigation of carbon-14 releases to the environment.

Neutron activation of other light elements is the principal mechanism by which natural and man-made carbon-14 come to exist. The contribution of carbon-14 from fission of reactor fuel is negligible (^{235}U fission yield is $1.7x10^{-6}$). The type of absorption and subsequent decay are factors, along with the nature of reactants and reaction medium, determining the chemical form newly created carbon-14 assumes. Following are nuclear reactions which can form the isotope in a LWR, listed with their respective thermal neutron reaction cross sections (σ_t):

Reaction	Importance	σ_t (barns)
$^{14}N(n, p)^{14}C$	Primary	1.81
$^{17}O(n, \alpha)^{14}C$	Primary	0.24
$^{13}C(n, \gamma)^{14}C$	Secondary	0.0009
$^{15}N(n, d)^{14}C$	Secondary	$2.4x10^{-7}$

Only the ^{14}N and ^{17}O reactions occur with great enough frequency to warrant consideration as sources.

There are two types of light water reactor and numerous individual units of each category. Each unit differs in size, control mechanism, and cooling subsystems, so some "average" is required for a generic discussion. There are several common features with respect to fuel. After the original core loading, assumed to be approximately 100 metric tons heavy metal (MTHM), it is assumed that one third of the fuel is removed and replaced each year in the manner customary during the equilibrium cycle. Therefore, the activation equation assumes 33.5 MTHM of fuel is irradiated for three years at a burnup of 33,000 MWt-days per MTHM and 33 percent thermal efficiency. This combination of burnup, fuel mass and efficiency result in one GWe produced in a year, the amount of electrical energy produced by our "average" unit of 1,250 MWe operating at an 80 percent capacity factor. These assumptions are believed to conservatively account for fuel conditions experienced in reactor designs. The neutron flux in both reactor types is assumed to be 5×10^{13} neutrons·cm^{-2}·sec^{-1} in the fuel and coolant. All thermal neutron reaction cross sections are multiplied by the factor 0.6 to account for 1/v variation of the two cross sections of interest. It provides a conservative bounding factor which prevents high values if the neutron energy distribution is not exactly thermal.

2.1.1 Annual production of carbon-14 in the fuel of LWR's

Now the activity of carbon-14 produced in the fuel elements of the "average" reactor may be calculated. Two reactions, $^{14}N(n, p)^{14}C$ and $^{17}O(n, \alpha)^{14}C$, are to be considered. In general, the activity produced due to neutron activation is:

$$A = \frac{dN}{dt} = N\sigma\phi - N\sigma\phi\, e^{-\lambda_d t_{irr}} \qquad (1)$$

$$= \frac{fmL}{M}\sigma\phi\,(1-e^{-\lambda_d t_{irr}}) \qquad (2)$$

where the first term on the right side of equation (1) describes growth of the product element, the second term describes the product's decay and

A = activity produced, disintegrations·sec^{-1}

f = fractional isotopic abundance of target element

L = Avogadro's number, 6.025×10^{23} mole^{-1}

σ = reaction thermal neutron cross section, barns (10^{-24} cm^2)

ϕ = local neutron flux, neutrons$\cdot cm^{-2} \cdot sec^{-1}$

λ_d = decay constant of product nuclide

t_{irr} = irradiation time

N = number of target element atoms

m = mass of target element, grams

M = atomic weight of target element, g$\cdot$gram-mole^{-1}

For the reaction $^{17}O(n, \alpha)^{14}C$ the following data apply:

$f = 3.7 \times 10^{-4}$

$m = 4.5 \times 10^6$ grams of oxygen (all isotopes)

[Note: in 33.5 MTHM of fuel, approximately 4.5 MT are oxygen]

M = 17 grams$\cdot$mole^{-1}

$\sigma = 0.24 \times 10^{-24}$ cm^2

$\phi = 5 \times 10^{13}$ neutrons$\cdot cm^{-2} \cdot sec^{-1}$

$$\lambda_d = \frac{0.693}{5{,}730 \text{ years}} = 1.21 \times 10^{-4} \text{ year}^{-1}$$

t_{irr} = 3 years

Therefore,

$$A = \frac{(3.7\text{x}10^{-4})(4.5\text{x}10^{6} \text{ g/GWe yr})(6.025\text{x}10^{23} \text{ mole}^{-1})(5\text{x}10^{13}\text{cm}^{-2}\cdot\text{sec}^{-1})}{(17 \text{ g}\cdot\text{mole}^{-1})}$$

$$\times \frac{(0.24\text{x}10^{-24} \text{ cm}^2)(0.6)(1 - \exp[-(1.21\text{x}10^{4})(3)])}{(3.7 \times 10^{10} \text{ sec}^{-1}\cdot\text{Ci}^{-1})}$$

A = 4.1 Ci/GWe-yr for the oxygen-17 reaction.

The activity produced in the activation of N-14 can be calculated in a similar fashion. The mass of nitrogen present is variable as it is a fuel impurity, and may be as high as 220 ppm. Rarely is the fraction greater than 20 ppm, which has been used for this calculation, and 20 ppm corresponds to about 756 g of nitrogen. Again, the data are listed:

$f = 0.99635$

$m =$ 756 grams of nitrogen

$M = 14$ grams $\cdot$ mole^{-1}

$\sigma = 1.81 \times 10^{-24}$ cm^2

$\phi = 5 \times 10^{13}$ neutrons$\cdot$cm$^{-2}\cdot$sec^{-1}

$$\lambda_d = \frac{0.693}{5{,}730 \text{ years}} = 1.21 \times 10^{-4} \text{year}^{-1}$$

$t_{irr} = 3$ years

Therefore,

$$A = \frac{(0.99635)(7.56\text{x}10^{2}\text{g/GWe}\cdot\text{yr})(6.025\text{x}10^{23}\text{ mole}^{-1})(5\text{x}10^{13}\text{cm}^{-2}\cdot\text{sec}^{-1})}{(14\text{ g}\cdot\text{mole}^{-1})}$$

$$\times \frac{(1.81\text{x}10^{-24}\text{ cm}^{2})(0.6)(1 - \exp[-(1.21\text{x}10^{-4}\text{yr}^{-1})(3\text{ yr})])}{(3.7 \times 10^{10}\text{ sec}^{-1}\cdot\text{Ci}^{-1})}$$

A = 17.3 Ci/GWe-yr for the nitrogen-14 reaction.

To summarize, carbon-14 is produced in the fuel by the two pathways at the approximate rate of 4.1 Ci/GWe-yr by the ^{17}O reaction and 17.3 Ci/GWe-yr by the ^{14}N reaction.

2.1.2 Carbon-14 produced in BWR coolant

The coolant of a light water reactor is exposed to neutron irradiation from the fuel elements. Water used for coolant is highly purified, though some residues remain dissolved or suspended which are subject to activation. Neutron absorption by O-17 bound as water should still predominate, however, because of the high target element concentration.

A representative General Electric BWR/6 contains 1,872 ft^3 of water pressurized to 1,062 psia and maintained at a temperature of 540°F. At these conditions, the water mass is 39.5 MT. The BWR/6 operates at a power level of 3,579 MWt, or an annual electric power output of 1.18 GWe. For this power level and water mass, carbon-14 is produced via the $^{17}O(n, \alpha)^{14}C$ reaction at the rate of 9.2 Ci/GWe-yr in the coolant.

No data was available on nitrogen concentration in the coolant of a BWR. The activity produced by a nominal 1 ppm of nitrogen is approximately 0.26 Ci/GWe-yr. It was concluded that this term be used awaiting more concrete data.

2.1.3 Carbon-14 produced in PWR coolant

The volume of coolant in a representative PWR (3,473 MWt, or 1.146 GWe at 33 percent thermal efficiency) was estimated from the core height of active fuel and cross sectional flow area of the core. A water mass of 13.7 MT was calculated from the specific volume at saturation conditions, 588°F. and under 2,235 psia pressure. The coolant is not saturated in the core, so the result may be somewhat low, though not greatly so. For these conditions, carbon-14 activity arising from the $^{17}O(n, \alpha)^{14}C$ reaction is produced at the rate of 3.3 Ci/GWe-yr.

Carbon-14 activity in the PWR coolant arising from N-14 neutron activation must be estimated as in Section 2.1.1.2. Data are not available on concentrations of nitrogen from air or control compounds, so a nominal value of 1 ppm was used. What results is a production rate of 0.09 Ci/GWe-yr.

All the results accrued in Section 2.1.1 are tabulated in Table 2-1.

2.2 Distribution of carbon-14 in the gaseous release pathways of a light water reactor

Carbon-14 is produced in the fuel and coolant and distributed wherever gas or fluid streams flow in the plant. Leakage of plant systems allows for eventual release to the environment, so the partitioning of original carbon-14 in various pathways is an important guide to the establishment of control measures.

Radioactive carbon will always be carried by stable carbon compounds. In boiling water reactors, much of the air entrained in the coolant is ejected from the main condenser overhead. This off-gas stream is fundamentally air and therefore carbon, as carbon dioxide, exists in approximately the same ratio to other constituents as it does in air. A very small amount of the stable carbon remains in the coolant; this level is probably regulated by chemical control. In a PWR, some CO_2 remains in solution as carbonic acid, while most leaks to an air space or

Table 2-1

Production Sources of ^{14}C in a Nominal 1000 MWe Light Water Reactor

Source	Reaction	Production Rate (Ci/GWe-yr)	Total (Ci/GWe-yr)
Fuel	$^{17}O(n, \alpha)^{14}C$ (0.037% abundance)	4.1	
	$^{14}N(n, p)^{14}C$ (20 ppm)	17.3	21.4
BWR Coolant	$^{17}O(n, \alpha)^{14}C$ (38.9 MT oxygen)	9.2	
	$^{14}N(n, p)^{14}C$ (1 ppm)	.26	9.5
PWR Coolant	$^{17}O(n, \alpha)^{14}C$ (11.5 MT oxygen)	3.3	
	$^{14}N(n, p)^{14}C$ (1 ppm)	0.09	3.4

to the chemical control system where it is diluted in nitrogen. Dilution in the off-gas streams is great enough to assure an air-like composition with respect to CO_2. Due to boric acid addition and buffering, a minor amount of CO_2 is expected to remain in the primary coolant. Corrosion control for the secondary system probably maintains a low CO_2 level, causing most to be released as gas in the air-ejector. Most critical, of course, are the stable carbon dioxide concentrations at the treatment point. These are provided where required in this chapter, Chapters 3 and 4 and the appendices.

2.2.1 Concentration of carbon-14 and potential magnitude of releases from BWR gaseous release pathways

The following systems have been considered as release pathways for gaseous carbon-14, and an effort has been made to estimate the magnitude of releases:

- condenser steam jet air ejector (SJAE)
- turbine gland seal condenser exhaust
- reactor building (including drywell) purge exhaust
- turbine building ventilation system exhaust
- radwaste building ventilation system exhaust

The concentration of stable carbon dioxide, compared on the basis of dry portions of vapor passing through these routes, is taken to be equivalent to that in air.

A cross-section of measured concentrations has been used when available. When these are lacking, the average carbon-14 concentration in the reactor coolant at Oyster Creek was utilized in conjunction with standard assumptions.(1-3)

Measurements at Oyster Creek indicate the gaseous releases of carbon-14 are a mixture of CO_2 and, presumably, organics. At the major release points, CO_2 dominates by at least two to one. The condenser steam-jet air ejector appears to be the most significant release point. Preliminary data from the Oyster Creek study(1) indicate an average C-14 release rate of 0.182 μCi/sec. On an annual basis, for a 1,250 MWe BWR operating at an 80 percent capacity factor, 9.0 Ci/yr is the estimated release. In a similar fashion, the release rate from the turbine gland seal condenser exhaust is estimated to be less than 0.015 Ci/yr. The latter figure is based on a measured release rate at Oyster Creek of $<3\times10^{-4}$ μCi/sec.

The latter three pathways must be calculated using measured coolant activities and leakage rates. The conservative assumption is made that all this leakage vaporizes and escapes to the building atmospheres. It is further assumed that the reactor and turbine building leakage is at 100 percent RCS activity, while that in the radwaste building contains only 1 percent of the RCS activity. The coolant concentration of carbon-14 at Oyster Creek is, on the average, 4.0 pCi/ml. Leakage rates are 500 lb/hr, 1,700 lb/hr and 1,000 gpd for the reactor building, turbine building and radwaste building, respectively. Resulting values are 1.1×10^{-2}, 3.8×10^{-2} and 5.5×10^{-5} Ci/yr for the reactor building purge exhaust, turbine building ventilation system exhaust and radwaste building ventilation system exhaust, respectively.

Results of Section 2.2.1 are tabulated in Table 2-2 and shown schematically in Figure 2-1.

Table 2-2

Summary of Gaseous Carbon-14 Releases from BWR Pathways

Pathway	Annual ^{14}C Release (Ci/yr)	Percent of Total
Condenser Steam-Jet Air Ejector Off-gas	9.0	99.3
Turbine Gland Seal Condenser Exhaust	0.015	0.2
Reactor Building Purge Exhaust	0.011	0.1
Turbine Building Ventilation	0.038	0.4
Radwaste Building Ventilation	5.5×10^{-5}	-
	9.1	

SOURCE	RADWASTE BUILDING VENT	REACTOR BUILDING PURGE EXHAUST	TURBINE BLDG. VENT	TURBINE GLAND SEAL CONDENSER EXHAUST	CONDENSER AIR EJECTOR EXHAUST
GASEOUS DISCHARGE FLOWRATE (SCFM)	50,000	10^6 FT^3, FOUR TIMES ANNUALLY	500,000	600	200
ESTIMATED ^{14}C RELEASE RATE (Ci/YR)	5.5×10^{-5}	0.011	0.038	0.015	9.0

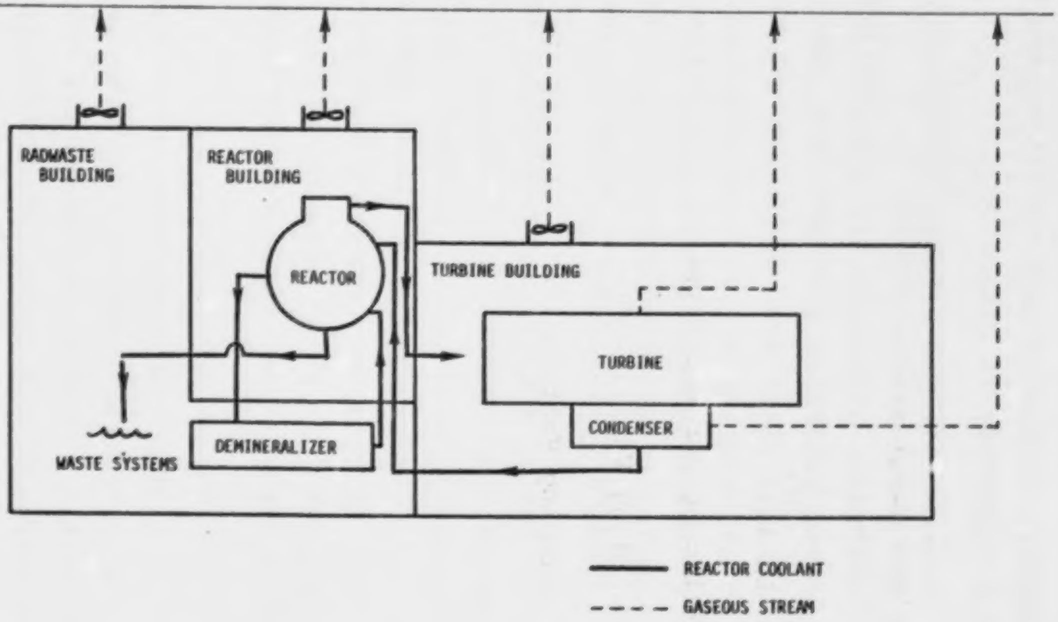
RADWASTE BUILDING
REACTOR BUILDING
REACTOR
TURBINE BUILDING
TURBINE
CONDENSER
DEMINERALIZER
WASTE SYSTEMS
REACTOR COOLANT
GASEOUS STREAM

Figure 2-1 Gaseous Effluent Pathways with Flowrates and Estimated Carbon-14 Releases for the Representative BWR

2.2.2 Concentration of carbon-14 and potential magnitude of releases from PWR gaseous release pathways

The following systems have been considered as release pathways for gaseous carbon-14, and an effort has been made to estimate the magnitude of releases:

- primary off-gas treatment vents (in normal and shutdown operations)
- condenser steam-jet air ejector (SJAE)
- steam generator blowdown tank vent exhaust
- turbine gland seal condenser exhaust
- fuel handling building ventilation exhaust
- containment ventilation system exhaust
- auxiliary building ventilation system exhaust
- turbine building ventilation system exhaust.

The concentration of stable carbon dioxide, where it is the dominant carbon compound in a pathway, compared on the basis of dry portions of vapor, is taken to be equivalent to that in air. Measurements of stable hydrocarbon concentrations, when hydrocarbon compounds dominate, are discussed below; in present form these data do not, in conjunction with standard assumptions, yield results having adequate certainty.

Once again an attempt has been made to use measured concentrations. When they are lacking, the average reactor coolant concentration of a representative PWR facility is used in conjunction with standard assumptions.

The chemical form of carbon-14 is different in releases from PWR's than one finds at BWR's. It was discovered that primary system off-gases emanating from the gaseous radwaste treatment system at Haddam Neck are virtually all in non-CO_2 form.(5) The CO_2 fraction of carbon releases from the condenser SJAE is similarly small, less than 15 percent at Haddam Neck.(5) Analysis performed for the Ginna PWR indicates nearly 90 percent of the gaseous radwaste system carbon-bearing off-gases are methane or ethane, whereas only 5 percent are CO_2.(4)

The most significant element contributing to carbon-14 releases is the off-gas stream from the primary gaseous radwaste treatment system. Flow and concentration data were studied to arrive at representative values which were in turn scaled to 1,250 MWe plant capacity if necessary. An average of waste gas decay tank carbon-14 concentrations, taken from Ginna, Haddam Neck, and Yankee Rowe and (4,5,6) was found to be 5×10^{-4} μCi/cm^3. Flow estimates range from 0.1 to 1.0 scfm in the off-gas system, supported by operating data for San Onofre 1, among others. That 430 MWe reactor processed 30,000 ft^3 in three months, which corresponds to 350,000 ft^3/yr for a 1,250 MWe plant. The scaled process rate from San Onofre, 0.66 scfm, generally supports an averaged flow of 0.5 scfm for the source term calculation. Resulting is an annual release rate of 3.0 Ci/yr from the gaseous radwaste system exhaust. Recall that all values reflect an 80 percent capacity factor.

Releases from the condenser steam-jet ejector exhaust are computed in a similar way. An average effluent concentration from Haddam Neck and Yankee Rowe was found to be 3.7×10^{-7} μCi/cm^3. Assuming 80 percent capacity factor in a 1,250 MWe PWR and a condenser SJAE flowrate of 25 scfm, one arrives at 0.11 Ci/yr projected release rate.

An estimate of steam generator blowdown tank (SGBT) effluent release rate was calculated with steam generator carbon-14 concentration data from Haddam Neck, 1.6×10^{-7} μCi/ml. Other conditions at that station include 5 gpm total blowdown flowrate, liquid flashing in the SGBT of 35 percent, and a further assumption that carbon-14 partitions in the same fraction as steam in the tank.(2) A release rate of 4.5×10^{-4} Ci/yr is predicted.

A conservative estimate for turbine gland seal condenser venting of carbon-14 must be made for want of operational data. It is assumed that 100 percent of the carbon-14 dissolved in the 50 gpd primary-to-secondary leakage partitions to the vapor phase and that 0.1 percent of the vapor phase (i.e., the main steam flow) is routed to the gland seal. Again, the average primary coolant concentration of C-14, observed at Haddam Neck and Yankee Rowe, is 1.67×10^{-5} μCi/ml. When these values are used to calculate the annual release rate of carbon-14 from the turbine gland seal condenser off-gas vent is 9.2×10^{-7} Ci/yr.

The concentration of carbon-14 in the Connecticut Yankee (Haddam Neck) plant's fuel handling building atmosphere was measured at 9×10^{-7} μCi/cm^3 of air. Using the operating ventilation rate of 70m^3/min on a continuous basis, the estimated release is 0.69 Ci/yr. Here again, the release rate was scaled to 1,250 MWe and an 80 percent capacity factor was assumed.

Measurements at the Ginna power station indicate a containment atmosphere carbon-14 concentration of 1.5×10^{-6} μCi/cm^3 of air during operation. The containment volume, assumed to be one million cubic feet, is purged four times annually. Consequently, it is estimated that 0.52 Ci/yr of carbon-14 is released from a 1,250 MWe PWR operating at an 80 percent capacity factor.

Calculations for the auxiliary building discharge rate based on measurements such as those taken at Haddam Neck are generally thought to be high for the reason that measured C-14 atmospheric concentrations are below the detection limit for C-14 in air, 6.0×10^{-9} μCi/cm^3. A reasonable alternative is to use the "standard" leak rate of primary coolant to the auxiliary building, equal to 20 gpd cold and 1 gpd hot, in concert with other system parameters to compute an estimate. Using the primary coolant C-14 average concentration of 1.67×10^{-5} μCi/ml, auxiliary building ventilation releases are estimated to be 8.0×10^{-4} Ci/yr.

Considerable uncertainty exists in some methods of estimating turbine building releases. The one utilizing a measured atmospheric concentration is most suspect because the value approaches the carbon-14-in-air detection limit. Other methods agree fairly well, so the one which yields conservative and reasonable results was chosen for this estimate. Secondary coolant in the steam generator of Connecticut Yankee has an average C-14 concentration of 1.6×10^{-7} μCi/ml. The secondary coolant is further assumed to leak to the turbine building at a "standard" rate of 1,700 lb/hr. At an 80 percent capacity factor in the nominal 1,250 MWe plant, an annual discharge rate estimate is 1.8×10^{-3} Ci/yr.

Results of Section 2.2.2 are tabulated in Table 3-3 and shown schematically in Figure 2-2.

Table 2-3

Summary of Gaseous Carbon-14 Releases from PWR Pathways

Pathway	Annual ^{14}C Release (Ci/yr)	Percent of Total
Gaseous Radwaste Treatment System	3.0	69
Condenser Steam-Jet Air Ejector Off-gas Vent	0.11	2.5
Steam Generator Blowdown Tank Vent	4.5×10^{-4}	0.01
Turbine Gland Seal Condenser Exhaust	9.2×10^{-7}	-
Fuel Handling Building Ventilation	0.69	16
Containment Building Purge Exhaust	0.52	12
Auxiliary Building Ventilation	8.0×10^{-4}	0.02
Turbine Building Ventilation	1.8×10^{-3}	0.04
TOTAL	4.32	

SOURCE	STEAM GENERATOR BLOWDOWN TANK VENT	GASEOUS RADWASTE TREATMENT SYSTEM	CONTAINMENT BLDG. PURGE EXHAUST	AUXILIARY BLDG. VENT	FUEL HANDLING BLDG. VENT	TURBINE BLDG. VENT	CONDENSER AIR EJECTOR	TURBINE GLAND SEAL CONDENSER EXHAUST
ASEOUS DISCHARGE FLOWRATE (SCFM)	1,200	0.5	10^6 FT^3, FOUR TIMES ANNUALLY	121,000	2470	100,000	25	500
ESTIMATED ^{14}C RELEASE RATE (Ci/YR)	4.5×10^{-4}	3.0	0.52	8.0×10^{-4}	0.69	1.8×10^{-3}	0.11	9.2×10^{-7}

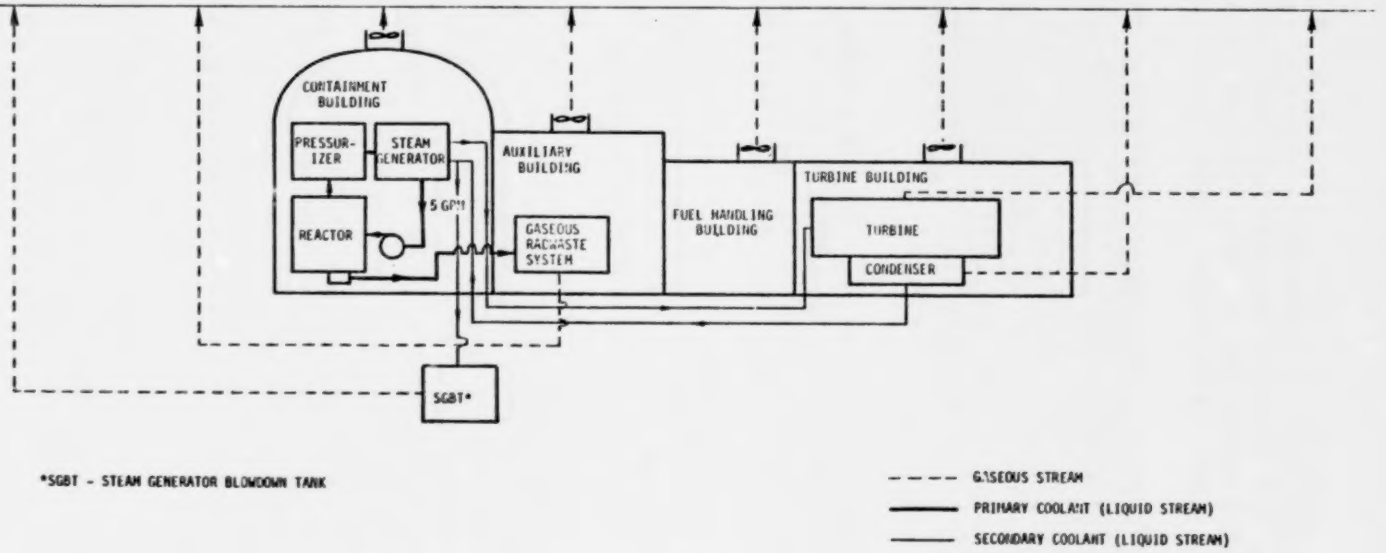
CONTAINMENT BUILDING
PRESSUR-IZER
STEAM GENERATOR
REACTOR
5 GPM
AUXILIARY BUILDING
GASEOUS RADWASTE SYSTEM
FUEL HANDLING BUILDING
TURBINE BUILDING
TURBINE
CONDENSER
SGBT*
*SGBT - STEAM GENERATOR BLOWDOWN TANK
GASEOUS STREAM
PRIMARY COOLANT (LIQUID STREAM)
SECONDARY COOLANT (LIQUID STREAM)

Figure 2-2. Gaseous Effluent Pathways with Flowrates and Estimated Carbon-14 Releases for the Representative PWR

Chapter 2 References

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CHAPTER 3. POSSIBLE C-14 CONTROL TECHNIQUES

Carbon dioxide is a gas that serves several commercial purposes: refrigerant, fire-extinguishing material, and intermediate for the manufacture of other chemicals. Because of the diverse industrial need for carbon dioxide, there are several techniques available for its collection and packaging. Selection of a specific process for the capture and retention of CO_2 depends on the volume of gas to be treated, the concentration in the gas stream, the composition of the gas stream, and the desired final packaged form.

Experience has shown that off-gas streams from nuclear power and reprocessing operations which may require carbon-14 treatment have approximately the same CO_2 concentration as that found in air.(20) Reference (dry) air has approximately the following composition:

	% by Volume
Oxygen	20.95
Nitrogen	78.08
Carbon Dioxide	0.03
Argon, etc.	0.94

The average molecular weight of air is 28.97 lbs/lb-mole.

Several processes which may have direct application to removal of carbon-14 from various reactor off-gas streams have been examined. Included in the examination were processes that have been designed specifically for noble gas removal. The nature and feasibility of each process is discussed in the following sections.

3.1 Scrubbing techniques

Scrubbing is a popular commercial method for the removal of one or more constituents from a gaseous stream. It involves absorbing the gaseous constituent that is desired to be removed into a liquid stream by passing the gaseous stream through the liquid.

Three scrubbing processes which allow for the removal of CO_2 from an air stream are scrubbing with water, aqueous caustic solution or ethanolamine.

3.1.1 Gas absorption by wet scrubbing

Carbon dioxide can be removed from an air stream by contacting the stream with water in a counter-current flow fashion using a packed absorption column. The process depends on normal gas absorption principles.

By studying the solubility of CO_2 in water in conjunction with the design procedure for packed towers, it was found that a tower of several hundred feet was needed to attain almost complete removal of the CO_2, which would be highly impractical. Space limitations in nuclear power plants and reprocessing facilities preclude this method. The attractiveness of such a system is further diminished because the final product (i.e., carbonic acid solution) would be in a liquid form which is unacceptable packaging for permanent isolation. Therefore, this process is not technically feasible for the desired application.

3.1.2 Caustic scrubbing by gas absorption

Absorption of CO_2 by scrubbing with a caustic aqueous solution is a familiar industrial process. Considerable attention is given in this section to caustic scrubbing as it represents the most probable candidate for actual application. Figure 3-1 is a flowsheet for a possible caustic scrubbing absorption system. The gas stream to be treated is passed through a blower and then introduced into a packed caustic absorption column. Carbon dioxide is stripped from the gas stream by the following chemical reaction:

$$2\ NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O$$

The bulk of the gas feed stream exists through the top of the column. It is filtered by at least one stage of roughing filters and one stage of HEPA filters before being released to the environment.

The scrubbing solution from the absorber column is batched in small lots to the mix tank when the desired conversion of CO_2 to Na_2CO_3 has been reached. Calcium hydroxide is then added to the mixture causing the carbonate to precipitate as calcium carbonate:

$$Na_2\ CO_3 + Ca(OH)_2 \rightarrow 2\ NaOH + CaCO_3$$

The amount of $Ca(OH)_2$ used to precipitate the $CaCO_3$ may have to be metered very carefully to avoid any excess which would cause saturation of the filtrate with $Ca(OH)_2$ (solubility is 2g/l) which in turn would react in the packed tower to form $CaCO_3$ which could plug the packing. Therefore, a direct caustic scrubber process using $Ca(OH)_2$ as the scrubbing agent may not be feasible.

The solution and precipitate are then pumped from the mix tank and filtered. The sodium hydroxide filtrate is cycled back to the absorber column, while the calcium carbonate cake is removed by the filter and packaged for final disposal as solid radwaste by incorporation into concrete.

The most important design aspect of caustic scrubbing systems is the sizing of the absorption column that will

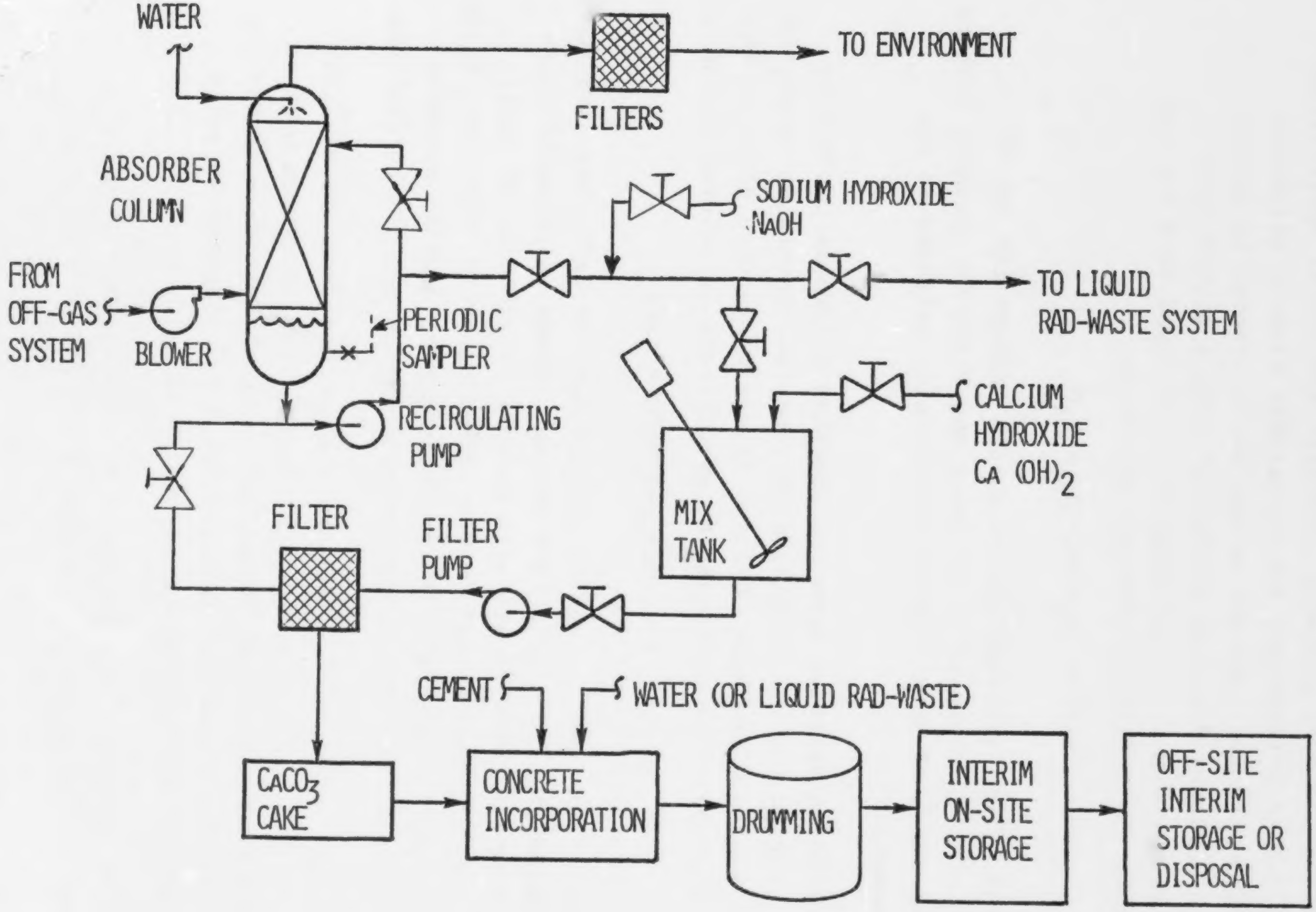
WATER
TO ENVIRONMENT
FILTERS
ABSORBER COLUMN
SODIUM HYDROXIDE NaOH
FROM OFF-GAS SYSTEM
BLOWER
PERIODIC SAMPLER
TO LIQUID RAD-WASTE SYSTEM
RECIRCULATING PUMP
CALCIUM HYDROXIDE Ca (OH)2
FILTER
FILTER PUMP
MIX TANK
CEMENT
WATER (OR LIQUID RAD-WASTE)
CaCO3 CAKE
CONCRETE INCORPORATION
DRUMMING
INTERIM ON-SITE STORAGE
OFF-SITE INTERIM STORAGE OR DISPOSAL

Figure 3-1. Flowsheet for a caustic scrubber to remove C-14

scrub the gas with sodium hydroxide. Design parameters for a gas absorption column include:

1) Gas flow rate and composition
2) Operating pressure and pressure drop across the absorber
3) Desired degree of recovery
4) Liquid flow rate
5) Operating temperature
6) Type of packing

The aqueous sodium hydroxide flow rate depends in each individual case on the desired air flow rate. The operating pressure of the absorber column will be maintained as close to atmospheric pressure as possible. The pressure drop across a packed column is a function of both liquid and air mass velocities. The limiting through-put factor of a caustic absorption tower is usually a flow condition that results in either foaming or flooding. Reference 1 provides empirical relationships between a scrubber solution that can be expected to cause foaming and ranges of both liquid and gas flow rates. A refernece design point obtained from these relationships is that an air mass velocity of 300 lbs/hour-ft^2 of column cross section can be passed through a packed column with a pressure drop of 0.5 inches of water per foot of packing height without causing major foaming problems. Flooding will occur in an absorber column containing a given packing and being irrigated with a definite flow of liquid if the gas flow rate exceeds some upper limit. The gas velocity corresponding to this limit is called the flooding velocity. The upper limit can be found from an inspection of the relation between the pressure drop through the bed of packing and the gas flow rate, from observation of the holdup of liquid, and by the visual appearance of the packing. The flooding velocity varies somewhat with the method of determination and appears more as a range of flow rates than as a sharply-defined constant. Reference 1 also provides empirical relationships between flow rates and pressure drops that indicate flooding conditions. Experience has shown that the most economical designs for caustic scrubbers utilize gas flow rates that are about 50-75 percent of flooding rates,(4) and pressure drops between .25 and .5 in. H_2O per foot of packing.(1)

Selection of the proper type of packing for the desired gas absorption operation is very important. The principal requirements of a column packing are:

1) The packing should be chemically inert to the fluids in the column.
2) It must be strong and durable but without excessive weight.
3) The packing should contain adequate passages for both streams without excessive liquid holdup or pressure drop.
4) It must provide good contact between liquid and gas.
5) The cost of the packing must be reasonable.

Ceramic saddles or rings of about an inch in size provide the requirements for packing in a caustic scrubbing absorber column.

Another requirement of economical absorber column design is that adequate distribution of liquid and gas be maintained throughout the packed section of the column. The first objective is to distribute the liquid as evenly as possible at the top of the column. Experience has indicated that, in smaller diameter columns (2 feet or less), even when distribution is almost perfect initially, coalescence of the liquid into streams will occur causing channeling. To control the channeling effect, the liquid should be redistributed periodically as it flows through the packing. This is achieved by inserting porous redistribution plates at vertical intervals, in the packed section, of 5 to 10 column diameters. The redistribution plates are designed to act as a coarse sieve, provide packing support, and yet not to inhibit the countercurrent flow of the gas. They are commonly referred to as gas injection support plates.

The recovery efficiency for an absorption tower can be predicted using a parameter known as the overall gas mass-transfer coefficient, $K_G a$, whose units are lb. moles/hr-ft^3-atm.(1) The overall gas mass-transfer coefficient for the column is analogous to the overall coefficient in heat transfer. It is determined by combining the effects of the local coefficients in much the same way as the overall heat transfer coefficient is derived from the individual heat transfer coefficients of the system. Thus, $K_G a$ is an overall measure of the resistance to mass transfer. $K_G a$ is based on a calculated overall driving force and can be represented by the following equation;(3)

$$K_G a = \frac{N}{h\ A\ \Delta P_{LM}}$$

where,

N = lb. moles solute material (CO_2) transferred per hour

h = height of packing, ft.

A = cross sectional area of the column, $ft.^2$

ΔP_{LM} = log mean partial pressure drive of the solute, CO_2

It is seen that K_Ga depends on the amount of gas to be absorbed, time required for passage, volume of the absorption column, and the operating pressure of the column. The mass-transfer coefficient for the absorption of carbon dioxide from a carrier gas using sodium hydroxide is approximately 2.25.(1) This is a conservatively low figure because there is some evidence that the value of K_Ga approaches infinity with small driving forces (e.g., less than 50 PPM soluble gas in the total). However, so little is known about the vapor pressure and therefore back-pressure in this region of small driving forces, that this is more or less speculative.

The removal of CO_2 by caustic scrubbing involves absorption accompanied by a chemical reaction. Experience indicates that use of a solution 2 N in sodium hydroxide and maintenance of a carbon dioxide to carbonate conversion of 15-25 percent will generally yield optimum absorption of CO_2.(5) Detailed cost estimates are provided in subsequent chapters.

3.1.3 Ethanolamine scrubbing

Gas scrubbing with ethanolamine ($HOCH_2CH_2NH_2$) was considered for the removal of carbon dioxide. This technology involves absorbing the CO_2 into an ethanolamine solution at ambient temperatures in a scrubbing tower. The solute is then steam stripped back out of the scrub solution in a second contactor. Such a process removes CO_2 from one gas stream and produces another somewhat richer gas stream in CO_2.

Past applications of this technology have been to streams where CO_2 has been at combustion product levels (>5 volume percent). In the present situation it exists at air concentration levels (.03 volume percent).

The particular operating problem of ethanolamine is its oxidization to corrosive oxalic acid and glycine, which has been experienced with such a process in the gas industry.(7) This would probably be amplified in a radiochemical application. The benefit of possible increased concentration of CO_2 in the gas is judged to be outweighed by the complexity and size of this system, and by the unknowns inherent in such a demanding application of relatively unsophisticated technology. A product solidification technique would be required in addition to the ethanolamine scrubber, resulting in the need for a caustic scrubbing column, although this column may be smaller. For all these reasons ethanolamine scrubbing was rated unfavorable with regard to the present needs.

3.2 Other absorption techniques

3.2.1 Lime bed absorption of CO_2

Lime is a widely-used compound in many industrial processes, including gas absorption and desiccation. Lime has been considered as a method to remove CO_2 from an air-like off-gas stream. The process is a gas absorption process accompanied by a chemical reaction. First, quicklime (CaO) is slaked by adding water, to form calcium hydroxide. Slaked lime prepared in this way is a white powder that can be mixed with more water and sand to form mortar. The mortar hardens by slowly taking up carbon dioxide and forming calcium carbonate.(8) Gas absorption in a lime bed is a limited application of this process. To avoid hardening of the bed, the lime absorber must be replaced fairly often, with only moderate buildup. This system does not represent an efficient use of materials. Large volumes of waste and the slowness of the chemical reaction make this method appear technically less feasible than caustic scrubbing for the required application of continuous or semi-continuous operation.

3.2.2 Absorption of carbon-14 dioxide in a fluorocarbon solvent, dichlorodifluoromethane

Considerable discussion has been devoted to this method of removal of hazardous radioactive off-gas constituents in recent years. Tentative preliminary designs have been proposed for reactor(9) and nuclear fuel reprocessing facility(10) off-gas systems. However, experience to date has centered on an experimental unit at the Oak Ridge Gaseous Diffusion Plant. Discussions of this system have been presented with respect to Xe-Kr removal experiments by Stephenson, et al.,(11-13) with respect to commercial reprocessing applications by Murbach, et al.(10) and most recently with respect to Kr-Xe and CO_2 removal by Stephenson and Eby.(14) Considerable evidence has accumulated using the Oak Ridge system; the following discussion is based primarily on that data.

Many gaseous products of reactor operation, among them carbon-14 as CO_2, are quite soluble in liquid refrigerant-12, dichlorodifluoromethane. Krypton, xenon and carbon dioxide are considerably more soluble than other volatile gases which might be in the effluent stream, and favorably temperature sensitive relative to the others. Differences in relative solubility make separation possible. Because lower temperatures increase the extraction efficiency, systems are designed to operate at cryogenic levels. In one system conceptualized for PWR application, a temperature of -27°F is chosen.(9)

One design has found pilot experience at Oak Ridge Gaseous Diffusion Plant.(9) When scaled down to a size compatible with a 1,000 MWe PWR, the design flowrate is 1 scfm. Feed gas is dried, compressed to the absorber column operating pressure, and passed through a molecular sieve to remove trace water, oil and other fouling agents (for this application CO_2 is considered a fouling agent). Then the gas is chilled to appropriate temperatures and passed to the absorption column for countercurrent extraction. The solvent is drawn off and stripped of dissolved noble gases, which can then be stored at 0.1 percent of the original feed gas volume. Solvent refrigerant-12 is chilled and recycled to the absorption column.

Tests at the pilot plant demonstrated a removal efficiency of 99.90 percent for krypton and 99.99 percent for xenon.(9) Total vented gas in this case contained about 0.1 percent of its input activity, or a decontamination factor of 1,000.

No design or demonstration has been found for this system on a BWR. The fundamental difference expected is the much higher flowrate experienced at the SJAE outlet. Any BWR fluorocarbon absorption system must have a much higher input capacity, though the principle is similar in all other regards.

The relative solubilities of various off-gas constituents are presented, in Figure 3-2, as Henry's Law constant versus temperature.(9, 14) It can be seen that carbon dioxide is more soluble even than xenon, one of the two noble gas fission products for which the fluorocarbon solvent technique was originally proposed. Carbon-14 is removed from the off-gas stream by low temperature sorption just as are krypton and xenon. A recent paper on the application of the refrigerant-12 off-gas removal technique to a reprocessing plant(14) indicates that CO_2 removals were higher, in tests, than those for xenon, as Figure 4-2 predicts. This poses a complication for a system intended to separate noble gases from the feed gas stream because carbon dioxide would concentrate in the solvent and decrease its activity during recycle. It is for this reason and others (fouling agents such as NO_2 and water) that a molecular sieve pre-treatment was suggested by Murbach, et al.(10) with respect to a potential design for application for the BNFP Separations Facility. In fact, carbon dioxide removal using this method requires one extra step, either pretreatment with a molecular sieve or an additional solvent stripping step. Nonetheless, this system merits consideration as a potential part of an integrated process. Cost estimates for the integrated process itself is outside the scope of this report, but those for the molecular sieve adsorption system as an independent C_{14} control method will be provided in Chapter 4.

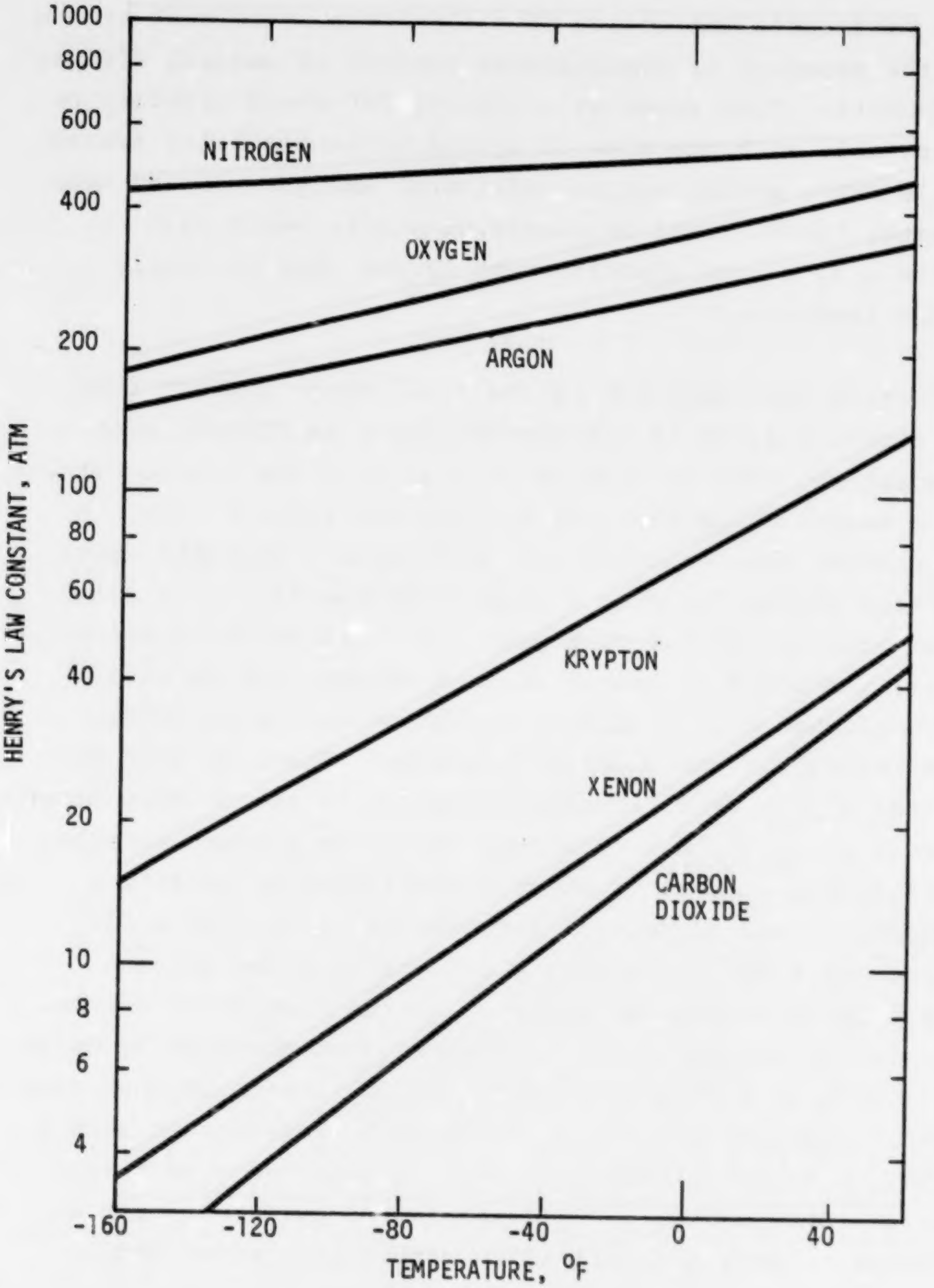

Figure 3-2 Henry's law constant as a function of temperature for dichlorodifluoromethane (Freon-12)

The most appealing aspect of a fluorocarbon solvent system is the prospect of simultaneous removal of several off-gas offenders. Flow rates at a current PWR would probably be compatible with the size of system currently being tested, while BWR's would require additional analysis due to much greater flows. Fuel reprocessing plants would also require a flow rate capacity much larger than currently under analysis.

A further consideration is the final waste product form. If carbon dioxide is not removed prior to fluorocarbon absorption, then it must be removed from the product stream in a manner which provides a permanent storage form: a compressed gas, which is not considered a suitable waste storage medium for such a long-lived nuclide, or a solid form such as calcium carbonate.(15) The latter alternative, discussed at length in this report, may be easily accomplished with a caustic scrubbing device in common use throughout the chemical industry. There is then the matter of CO_2 which remains dissolved in the solvent after normal stripping has occurred. Prior to solvent recycle, further CO_2 removal might be accomplished by passing a gaseous stream of solvent refrigerant-12 through a 13X molecular sieve, following which the purified solvent could be condensed and returned to the absorption column. The use of 13X molecular sieves has been shown to be quite effective in purifying R-12.(15) Experiments in that same study indicated the 13X sieve could be regenerated with a 350°F, 6.85 cfm nitrogen purge flow supplied over 6 to 7 hours. Once again, $^{14}CO_2$ contaminated purge gas must be treated to create a solidified carbon-14 product form.

It is concluded that absorption in a fluorocarbon solvent is a potential carbon-14 removal technology. It presents a possible method for simultaneous removal of several off-gas constituents currently being reviewed as potential health hazards. Such a system also has disadvantages in this instance:

- passage of off-gas containing carbon-14 dioxide through this system results in multiple streams requiring removal of lesser amounts of the original CO_2 gas volume
- achievement of a final solidified product form will require further treatment of gaseous product streams emanating from a fluorocarbon extractor
- a carbon-14 dioxide contaminated fluorocarbon solvent will be created requiring waste management consideration.

These disadvantages must be weighed against the advantages of such a system when a more specific design becomes available. Perhaps it will be found to be more cost-effective to remove CO_2 prior to fluorocarbon (or another technique) noble gas removal by a system which provides a one-step solidified product such as caustic scrubbing. In any event, final judgment must await more specific data than currently available and balancing of potential techniques to accomplish cost optimization.

3.3 Adsorption techniques

Adsorption on a solid medium is another popular method for the removal of one or more constituents from a gaseous stream. The solid medium is usually arranged in a fixed bed configuration with the gaseous stream passing through the bed. The gaseous stream enters the bed, the constituent which is to be removed is adsorbed on the solid medium and the gaseous stream exits leaving the removed constituent behind.

3.3.1 Fixed bed adsorption of carbon dioxide with molecular sieves

Another unit operation that can be used to remove carbon dioxide from a gas stream is fixed bed adsorption. Fixed bed adsorption is widely used to remove small quantities of water, hydrogen sulfide, and carbon dioxide from other gases. It is used primarily as a pre-treatment of feed stock to cryogenic processes such as air liquefaction and the manufacture of liquified natural gas.(2)

The capture of carbon dioxide by fixed bed adsorption using molecular sieves would require a pre-adsorption step to dry the feed gas stream, removing essentially all of the water. Two dessicant beds would be used so that one bed could be on line removing water from a continuous flow of gas while the second bed was being regenerated by a hot air purge. Purge gas from the bed being regenerated would be passed through a condenser and most of the captured water would have to be routed to the liquid rad-waste system. The dried feed gas stream is then routed through a molecular sieve bed for the removal of carbon dioxide. As in the case for the desiccating beds, two fixed beds would be needed so that one could be receiving a continuous gas supply while the other was being regenerated. A widely used molecular sieve that effectively removes CO_2 is sodium zeolite.

The bed being loaded would have to be held between -75° and -78°C to achieve good adsorption. Bed temperature is the most important parameter in loading considerations.

The regeneration of a loaded bed is accomplished by heating it to a temperature between 150° and 350°C, then

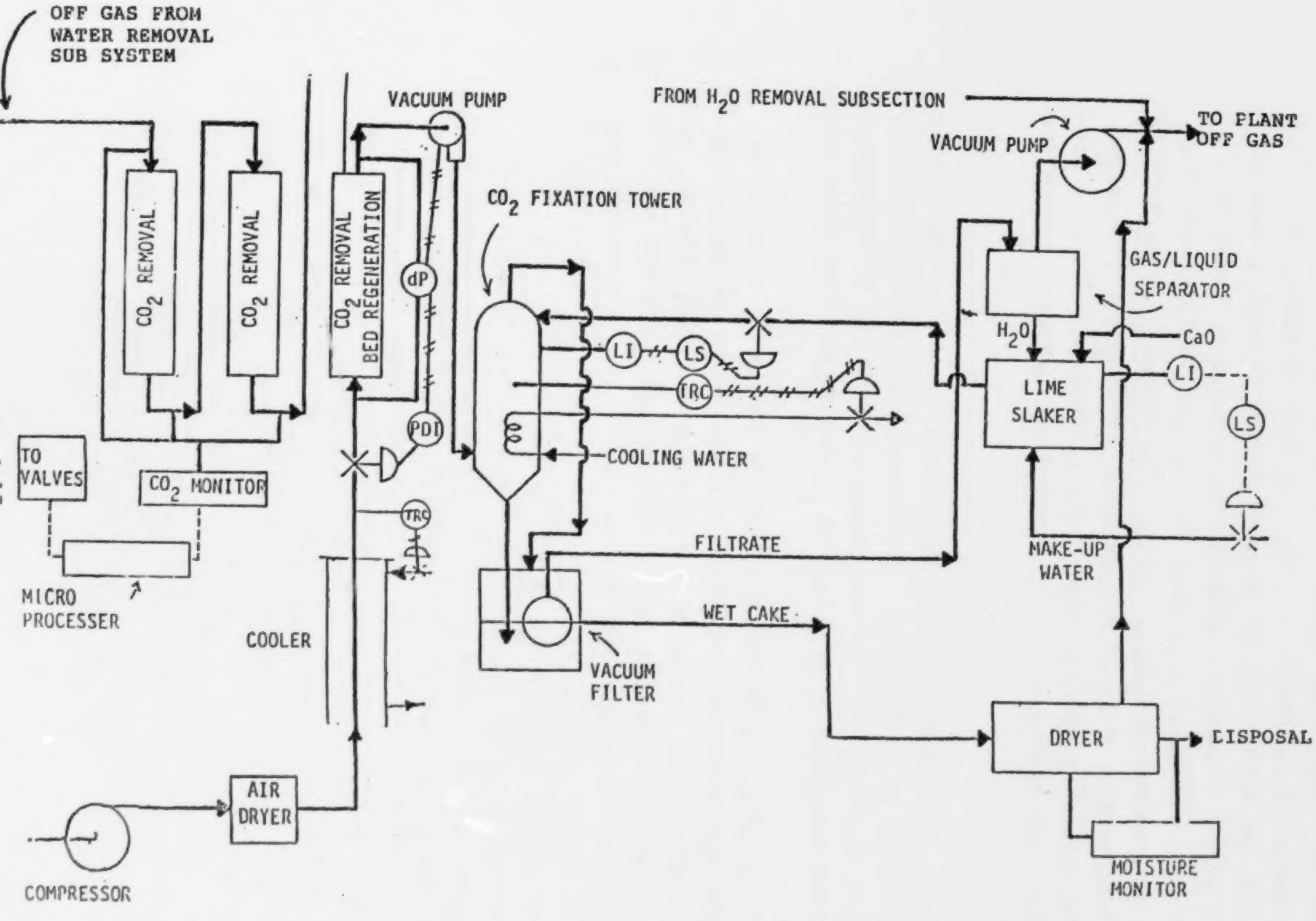
OFF GAS FROM
WATER REMOVAL
SUB SYSTEM
CO_2 REMOVAL
CO_2 REMOVAL
CO_2 REMOVAL
BED REGENERATION
VACUUM PUMP
dP
PDI
TRC
TO VALVES
CO_2 MONITOR
MICRO PROCESSER
COOLER
AIR DRYER
COMPRESSOR
CO_2 FIXATION TOWER
LI
LS
TRC
COOLING WATER
FILTRATE
WET CAKE
VACUUM FILTER
FROM H_2O REMOVAL SUBSECTION
VACUUM PUMP
TO PLANT OFF GAS
GAS/LIQUID SEPARATOR
H_2O
CaO
LIME SLAKER
LI
LS
MAKE-UP WATER
DRYER
DISPOSAL
MOISTURE MONITOR

FIGURE 3-3 CO_2 REMOVAL SUBSYSTEM PIPING AND INSTRUMENTATION.

passing a gas purge through the bed at the elevated temperature to remove the CO_2, as show in Figure 3-3(16). Purge gas would have to be next passed through a caustic scrubbing column or a slaked lime process in order to isolate carbon-14 in a form that is currently acceptable for permanent disposal. Again, the carbon dioxide is precipitated as calcium carbonate. Following filtration, the calcium carbonate cake would be incorporated into concrete and packaged for interim storage and/or ultimate disposal.

There is concentration of CO_2 before it enters the caustic scrubber or lime slaker. A cost estimate for this combination of molecular sieve-scrubber is provided in Chapter 4, along with that of a molecular sieve-slaked lime combination.

The possibility of using molecular sieves in a throwaway mode has also been examined. It would involve adsorbing the CO_2 on the sodium zeolite medium and then removing and packaging the medium rather than regenerating the bed. This technique has been deemed technically infeasible for the reason that a large fraction of the CO_2 trapped in the sieves is desorbed as the temperature returns to ambient from the cryogenic operating temperatures. Once again, if this product could be physically isolated, it is felt a gaseous form is unsatisfactory (the product must be considered to be in a gaseous form even in an adsorbed condition).

3.4 Control of carbon-14 using an integrated cryogenic distillation technique

One alternative to storage of gaseous reactor products is cryogenic separation. This principle depends on differences in boiling points of the several off-gas constituents. At temperatures achieved with a liquid nitrogen coolant, the various gases fractionate as follows:

Liquid, Solid or Slush	Gas
Xe (B.P. = -107°C)	N_2 (B.P. = -196°C)
Kr (B.P. = -153°C)	O_2 (B.P. = -183°C)
CO_2 (B.P. = -79°C)	CO (B.P. = -190°C)
	H_2 (B.P. = -253°C)

(All boiling points are for one atmosphere cover pressure)

Some constituents of the gas stream are liquified, fractionally distilled and the distillates collected. Typical purity limitation is encountered due to vapor pressures of the liquid components,* impurities in the reactor off-gases from which the liquor is composed and radiolytic products formed in the distillation apparatus. Nonetheless, product gases become separated to a high degree and they can be stored at a greatly reduced volume and subsequently released following decay holdup. They might also be bottled and stored for long-term decay. The latter alternative provides for nearly zero release of noble gas, while both processes effectively remove iodine. In either case, decontamination factors measured from the SJAE off-gas stream to the purified off-gas vent are taken to be 10,000 and 2,500 for Xe and Kr, respectively.(17) It is estimated that the same DF is achieved by about 20-40 days of charcoal holdup assuming the liquid effluent is held and released, and not bottled. One vendor warrants its product for a krypton DF of 10,000. (18)

---

*Approximately 0.01 percent of the Kr and iodine and 0.025 percent of the Xe are vented with the carrier gases.

There has been little experience with cryogenic off-gas systems on operating reactors, but designs have been tested in many liquid air separation and other purification processes. Appurtenances and feed/output possibilities vary in different designs. In one choice, feed gas can come directly from the SJAE line and pass through a hydrogen-oxygen recombiner, holdup line and gas dryer prior to entry in the distillation loop. However, to allow for continuous operation during shutdown of the distillation apparatus, a holdup tank or redundant distillation device is provided, whichever is most economical. Some holdup time in the egress line prior to venting provides decay time for nitrogen and other activation products created in the recovery and separation columns.

The distillation apparatus are similar in each system reviewed. Feed gas is deoxygenated to prevent excessive ozone formation. It is then dried and chilled in a regenerative pre-cooler to remove any high boiling components which would clog the recovery column or reduce its efficiency. The gas stream then passes, in a counter-current fashion, in contact with liquified nitrogen (LN_2) coolant in which all products except nitrogen ozone, traces of oxygen and hydrogen, and carbon monoxide are liquified. The vent gases, which now contain very little Kr-85, are passed through a regenerative precooler and vented to the atmosphere. The liquid fraction passes to a separation column where it is fractionally distilled and the vaporized constituents are removed and held separately. Alternatively, the liquified products may be left as a mixture and stored for decay in a holdup tank, then released after 45-90 days (the time period is only for illustration). If bottling is employed, on-site shielded remote handling facilities must be available for the Kr-85 fraction until it is packed in a transfer cask. A further design feature on some separation columns is a recycle line to the feed gas holdup tank for any occluded or entrained radioactive species. The cryogenic apparatus is packaged in a positive pressure inert atmosphere to prevent leakage hazards.

Several BWR's have proposed the use of cryogenic distillation systems for their SJAE off-gas treatment to accomplish noble gas fission product decontamination.(19, 20)

Carbon dioxide has generally been regarded to be a fouling agent in most systems reviewed because of its relatively high freezing temperature. The freezing point of carbon dioxide is also a triple point, so standard liquid to vapor fractionation cannot normally be performed. Freezeout of CO_2 in the liquid nitrogen absorber or the distillation apparatus can impede heat exchange and flow streams. For these reasons, it has generally been planned to provide either a pre-cooling (initial freezeout) step as a cold-trap for CO_2 or parallel molecular sieves with regeneration capabilities. In either case, gaseous $^{14}CO_2$ must be treated in a manner providing a stable, solid final form, such as caustic scrubbing.

Molecular sieves could provide significant CO_2 concentration, so the use of molecular sieves with a cryogenic distillation unit provides some advantage for an integrated control system. On the other hand, if a cold-trap could be designed to remove the CO_2 before entry into the cryogenic distillation system, and the removal of the CO_2 from the cold trap could be done in a way that would concentrate the CO_2, a gas feed richer in CO_2 could be sent to the scrubber, thus reducing feed volumes and consequently the size of the scrubbing column. This is speculation, however, as such a method has not been developed and demonstrated for the commercial use described here.(21) Research and development efforts, as well as demonstration, would be needed before a technical and economic assessment could be completed. For all these reasons, cryogenic distillation is seen as a potential integrated removal technology for several radionuclides and not considered as a C_{14} removal method.

3.5 Summary

Several possible carbon-14 control technologies have been examined. Cost estimates will be provided for comparison purposes.

A cost effective system to be used for carbon-14 isolation will be designed, produced and maintained according to quality specifications. In actual practice, a reliability of 95% can be expected and required, with quality assurance programs providing documentation in support of activities controlled to that end.

Probable reliability of a system is an implicit consideration subsumed in statements regarding technical feasibility. A young technology is less favored than a mature one because operating information is lacking. The decision to use an untried system involves more risk, and that risk is one consideration of several reflected in recommendations favoring or disfavoring specific isolation technologies.

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21. Personal communications with nuclear reactor vendors and nuclear power plant owner/operators.

CHAPTER 4. MODIFIED LWR EFFLUENT TREATMENT SYSTEMS FOR THE CAPTURE OF THE MAJORITY OF C-14

The following independent C-14 control alternatives will be examined.

1. Caustic scrubbing
2. Molecular sieve
3. Molecular sieve/caustic scrubbing

In the integrated control cases which include cryogenic distillation or fluorocarbon absorption, CO_2 must be removed first by one of the above alternatives. Therefore no cost analysis will be given. NO_x removal being necessary for both C-14 removal and krypton removal, will be considered a sunk cost for both, resulting in some savings for the C-14 removal system. These savings will be addressed.

4.1 BWR off-gas C-14 treatment system

As indicated in Chapter 2 it is estimated that approximately 99 percent of the C-14 released from a boiling water reactor is via the steam jet air ejector exhaust (SJAE). The SJAE flow rate is approximately 200 cfm untreated and drops to between 20 to 30 cfm after being passed through an off-gas recombiner, condenser, and dryer system. The composition of the processed stream is essentially the same as air.

Many existing BWR's have off-gas recombiners as part of the basic reactor effluent treatment system. Those which do not have them may be required to retro-fit them, because they reduce the volume of gases to be delayed in the off-gas systems, thereby increasing decay time and reducing the quantities of radioactive gases released to the environment.(1) For this reason, the capital and annual costs of a recombiner system will not be included in this evaluation as part of a C-14 control system.

A preliminary economic evaluation will be performed to identify the most cost effective alternative.

4.1.1 Caustic scrubber system

As indicated in Chapter 3, caustic scrubbing is a plausible means of removing carbon-14 from BWR off-gas systems since almost all the C-14 is in the form of CO_2 in an air stream. Therefore a more detailed treatment is given to this method, although a range of cost estimates will be provided for the other alternatives which are technically feasible.

From Figure 3-1, it can be seen that the major equipment components of the scrubber include:

- off-gas feed blower
- absorption column and internals
- mix tank
- filter pump
- recirculation pump
- $CaCO_3$ filter

In addition to these major equipment items, supporting equipment is required including:

- piping
- valves
- instrumentation
- electrical supply and control systems.

Like the recombiner/condenser, the liquid radwaste solidification unit is considered to be part of the basic reactor system and is not part of the cost estimate. Such a solidification unit is a standard item and is required in all nuclear plants. It is expected that the majority of solidification units could handle the additional radwaste created by a C-14 control system.

Off-gas feed blower

The operating range of the feed blower is between 20 and 60 CFM with a ΔP of 20 inches of water. For purposes of this evaluation, it is assumed that the blower is operating at 40 cfm. A 3/4 HP direct drive blower is adequate. It will have a 230/460 - 3 phase motor. (4)

Additional equipment includes a surge tank, measuring pot, recirculation pump (1 HP, 40 feet of head), valves and piping.(5)

Carbon dioxide absorption packed column

The design procedure for the absorption column is presented in Appendix A. The overall length and diameter of the absorption column is dependent on the desired CO_2 removal. Table A-1 summarizes the results of Appendix A. The overall length of the column is 13.12 feet for 90 percent removal of CO_2, and 21.22 feet for 99 percent CO_2 removal. All column designs accept a gas feed flow of 40 cfm. This requires a column diameter of 12 inches for 90 percent and 99 percent removal of CO_2. The bottom two feet of the column act as a reservoir for scrubber solution (2 N NaOH). The top three feet of the column, which are above the packed section, serve as a disengaging section. Within this disengaging section is a deentrainment unit constructed of woven metal mesh 10 inches thick.(7) Above the deentrainment unit is a full cone, narrow angle (30°), 304 stainless steel, water wash-down nozzle which can be used to back wash the unit.(8) The column is fabricated with 1/4 inch 304 stainless steel.

For the purposes of this evaluation, the column is designed with 1 inch ceramic Berl saddles as packing. Any other reasonable packing could have been used. The packing is supported and gas feed is accomplished with gas injection support plates, and the liquid feed to the column is by means of a distributor to assure a good flowpath.(6)

The scrubber solution is recycled externally to the absorption column through a recirculation loop containing a recirculation pump. The pump is capable of pumping 30 feet of head and has the following specifications:

1 HP, 230/460 motor (10)
1 1/2 inch suction
1 inch discharge
6 inch impeller
Mechanical Seal

Mix tank

The mix tank has a 9 gallon receiving capacity. Nine gallons represents 25 percent of the scrubber column holdup plus 1 gallon of calcium hydroxide solution with a 20 percent freeboard still available. Such a vessel could be approximately 1 foot in diameter and 1.5 feet tall and fabricated from 1/4 inch, 304 stainless steel. The mix tank is also equipped with an agitator which is mounted on top of the tank with the propeller shaft entering the tank through a teflon lipped seal. The propellor and shaft are 304 stainless steel.(9)

$CaCO_3$ filter and filter pump

Two types of filter can be used to trap the $CaCO_3$. The first type is a 5-micron ethylene-propylene cartridge filter. The loading capability is about 35 grams per cartridge. The filter casing is made of 304 stainless steel and is capable of housing 18 cartridge filters.(14) The filters would have to be changed about once every 7 hours (once a shift).

Until recently, cartridge filters have been found to give the most efficient and economic service among radioactive waste filtration alternatives. One drawback of the cartridge filter is the volume of radioactive waste that is added to the system when the cartridges are discarded.

A second type of filter that has recently been introduced into the industry is an etched disk backflush filter. The filter is made with 316 stainless steel and has a loading capacity of 500-2,000 grams. It can be back washed with about 3 gallons of water.(13) Although this type of filter involves a much greater initial capital cost, the savings in not having to replace cartridges and decreased volumes of radwaste make it competitive in the long run.

The filter pump is similar to the recirculation pump discussed previously.

Other types of filters can also be used. The operating and waste disposal costs will be similar to those for the above reference cases. Therefore, they are not included in the subsequent economic analysis.

Piping and valves

The valves are two inch, 316 stainless steel air-operated and teflon sealed. They can be operated from the control panel.(10) There are eight valves as indicated in Figure 3-1.

The off-gas enters and leaves the scrubber system through 1/2 inch, 304 stainless steel, schedule 40 pipe. All system piping carrying liquid will be two inch, 304 stainless steel, schedule 40 pipe. All piping is of fully welded construction.(11)

Instrumentation and control systems

The instrumentation is basically pneumatic in nature with electronic read-out. All of the recorders and indicators read out on a local remote panel board. The following list of instruments will be required for the caustic scrubber system:

Equipment Item	Instrument
Off-gas feed blower	On-off light indicator
	Gas flow rate recorder
Absorption columns	Scrubber solution level recorder
	Temperature and pH recorder
	Recycle loop flow indicator

Equipment Item (continued)	Instrument (continued)
	Recirculating pump on-off light indicator
	Differential pressure indicator for packed section
	Differential pressure indicator across the deentrainment unit
	Deentrainment unit spray nozzle rotometer
Mix tank	Liquid level recorder
	Agitator on-off light indicator
Filter pump	Pump on-off light indicator
Filter	Differential pressure indicator

Cost Evaluation

Fabricated cost of equipment was used as the basic building block to estimate the total capital cost of the various C-14 cleanup systems. The majority of the equipment cost data was obtained by direct contact with vendors, fabricators, and construction firms.

It is assumed that the fabricated equipment costs represent 13.3 percent of the total capital cost, including both direct, indirect costs and contingency. A discussion on capital cost estimation and various cost factors can be found on pages 25-12 through 25-22 of Chemical Engineering Handbook, Edition Five by Perry and Chilton. Figure 4-1 outlines the capital cost estimating module concept utilized for this study.

The indirect costs shown in Figure 4-1 include engineering, normal contingency, contractor fees, construction overhead, administration, QA/QC, licensing fees and interest on capital during the construction phase. To the above direct and indirect costs, a 50 percent nuclear contingency is added to cover the more stringent "tightness" requirements for a nuclear facility.

The special facilities requirements such as thicker walls encountered in a nuclear reprocessing plant will be added to the capital costs where appropriate. For PWRs and BWRs, the concrete and steel factors shown in the module are sufficient, as no extra shielding is required for the C-14 control system.

apital Cost Estimating
Module Concept

	Direct Material, M (E+M)		Direct Labor, L (L)		Direct Costs ((E+M+L)
Fabricated Equipment	100				
Piping	43				
Concrete	10				
Steel	5		85		
Instruments	18				
Electrical	9				
	185	+	85	=	270

↓

Indirect
Cost Factor
(X1.85)

↓

Direct and Indirect Cost Factor	500
Nuclear Contingency	250
Total Module Cost Factor	750

Figure 4-1

The annual carbon fixation costs for the various C-14 cleanup systems are calculated as present worth values assuming a 30-year operating life for a reactor and an 8 percent compound interest rate. The operating costs include labor, chemicals, maintenance and replacements. Annual fixed charges are 20 percent of capital costs.

Fabricated equipment costs for a BWR caustic scrubber removal system for C-14

Table 4-1 summarizes the fabricated equipment costs for a BWR off-gas C-14 removal system. Two system options are addressed:

Option A - The system contains disposable cartridge filters.

Option B - The system contains a backflush filter.

Within each option are defined the following cases:

Case I - 90% removal of entering CO_2
Case II - 99% removal of entering CO_2

Table 4-1

Fabricated Equipment Costs for BWR C-14 Scrubbing System

Item		Cost
1. Off-Gas Blower		$ 490
2. Absorption Column	- Case I	2,400
	- Case II	3,900
3. Column Internals		
a. Packing	- Case I	90
	- Case II	180
b. Gas Injection Support Plates		
	- Case I (1)	210
	- Case II (2)	420
c. Liquid Distributor		
	- Case I	220
	- Case II	220
d. Wire Mesh		25
e. Wash Down Nozzle		36
4. Mixing Tank		360
5. Agitator		500
6. Chemical Charging Tanks		
a. NaOH		350
b. $Ca(OH)_2$		350
7. Recirculation Pump		1,200
8. Filter Pump		1,200
9. $CaCO_3$ Filter Unit		
Option A - cartridge filter		1,300
Option B - backflush filter		10,000

Table 4-1 (continued)

Item	Cost
Total fabricated equipment cost	
- Option A	
a. Case I	$ 9,000
b. Case II	10,000
- Option B	
a. Case I	$17,000
b. Case II	19,000

Based on the above discussion, the total capital cost of these systems can be expected to be as follows:

Option A	
Case I	$ 67,500
Case II	75,000

Option B	
Case I	$128,000
Case II	143,000

Annual CO_2 fixation costs for a BWR off-gas C-14 treatment system by caustic scrubbing

The total labor costs associated with the operation and maintenance of a carbon-14 control system are spread across a reactor facility's entire organizational structure. This additional off-gas treatment unit requires support from the maintenance, engineering, QA/QC, and administrative staff.

Figure 4.2 is a typical reactor staffing plan.(18) This diagram indicates the complex interactions of a reactor staff. Figure 4.3 shows a second staffing plan.(19) The personnel requirements for Figure 4.3 are summarized in Table 4.2.

The operating staff of a reactor plus the supporting administrative staff number nearly 100 people. It is anticipated that the additional work load for a reactor staff from a retrofitted Carbon-14 control system would be approximately one percent of the total facility work load. Depending on how cost-effective the staffing plan is at a given site, staff numbers may change. As an example, if the staffing has some excess man power at most points within the organization there may not be a staff increase. But if at a specific reactor a shortage of staffing exists, more than one staff member may be required.

It is estimated that the general quality of staffing addition required for the approximately one percent work load increase would be at the operator qualification level. A new reactor with a carbon-14 control system incorporated into the basic design may have a slightly reduced additional work load, but not reduced significantly. The annual operating costs for the systems include labor, chemicals, utilities, maintenance, and replacement. Maintenance and replacement costs include repair and replacement of equipment. For this study, 7 percent of the original capital cost is assumed for the maintenance and replacement costs.(17) An average capital cost of $100,000 is assumed for the above options. The labor cost assumes one additional staff member at the qualification level of an operator at $25,000 per man year. An illustration of operator costs can be found in Figure 4-4. Chemical costs are assumed to be $1,000 per year and utility costs are negligible. The time requirements to operate the system with cartridge filters is the same as for the system with back flushable filters. In one case an operator is removing filter cartridges, and on the other the operator is running a control panel. In both cases attendance during operation is required, whatever be the activity of an operator during that time. Therefore, the operating costs are:

Labor		
$25,000 x 1.0	=	$25,000
Maintenance and Replacement		
$100,000 x 0.07	=	7,000
Chemicals		1,000

An annual fixed charge (20% of the capital costs) results from insurance, taxes and depreciation.

Fixed charges	$20,000
Total annual costs of fixing CO_2 as $CaCO_3$	$53,000

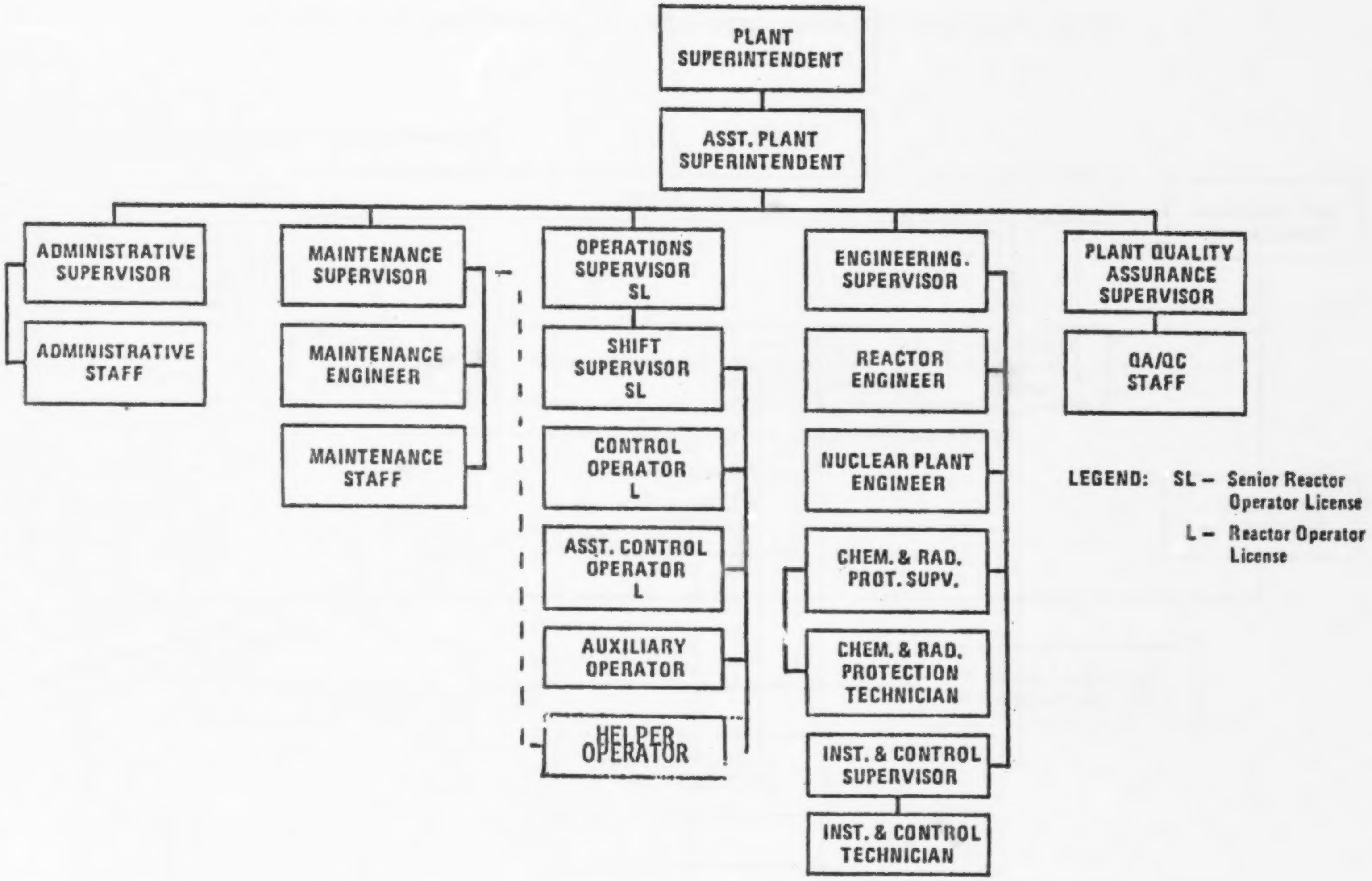
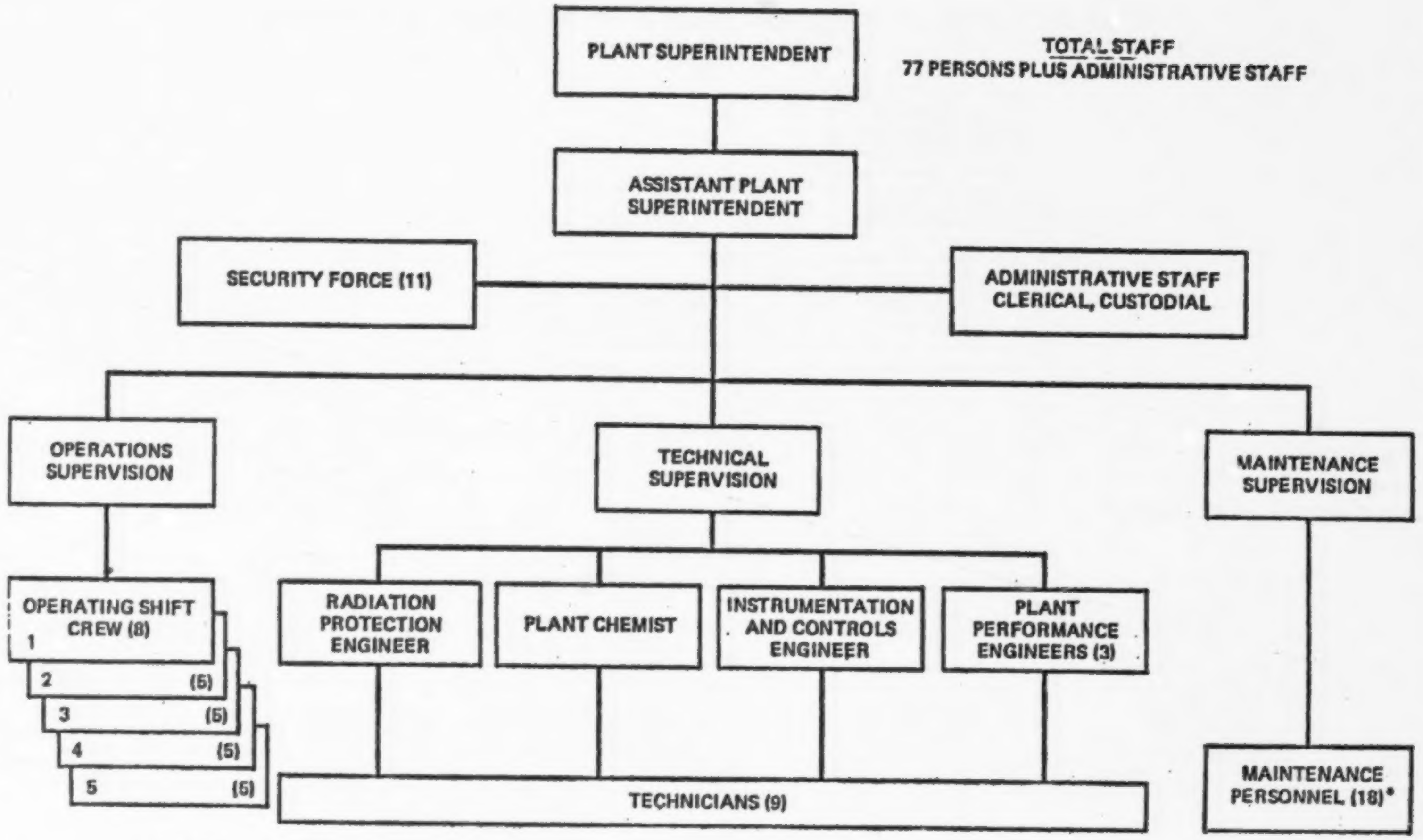

PLANT SUPERINTENDENT
ASST. PLANT SUPERINTENDENT
ADMINISTRATIVE SUPERVISOR
ADMINISTRATIVE STAFF
MAINTENANCE SUPERVISOR
MAINTENANCE ENGINEER
MAINTENANCE STAFF
OPERATIONS SUPERVISOR SL
SHIFT SUPERVISOR SL
CONTROL OPERATOR L
ASST. CONTROL OPERATOR L
AUXILIARY OPERATOR
HELPER OPERATOR
ENGINEERING. SUPERVISOR
REACTOR ENGINEER
NUCLEAR PLANT ENGINEER
CHEM. & RAD. PROT. SUPV.
CHEM. & RAD. PROTECTION TECHNICIAN
INST. & CONTROL SUPERVISOR
INST. & CONTROL TECHNICIAN
PLANT QUALITY ASSURANCE SUPERVISOR
QA/QC STAFF
LEGEND: SL – Senior Reactor Operator License
L – Reactor Operator License

Figure 4.2: Example of a Typical Reactor Staffing Plan


PLANT SUPERINTENDENT
TOTAL STAFF
77 PERSONS PLUS ADMINISTRATIVE STAFF
ASSISTANT PLANT SUPERINTENDENT
SECURITY FORCE (11)
ADMINISTRATIVE STAFF CLERICAL, CUSTODIAL
OPERATIONS SUPERVISION
TECHNICAL SUPERVISION
MAINTENANCE SUPERVISION
OPERATING SHIFT CREW (8)
1
2 (5)
3 (5)
4 (5)
5 (5)
RADIATION PROTECTION ENGINEER
PLANT CHEMIST
INSTRUMENTATION AND CONTROLS ENGINEER
PLANT PERFORMANCE ENGINEERS (3)
TECHNICIANS (9)
MAINTENANCE PERSONNEL (18)*

*Augmented by 10 special craft located off site

Figure 4.3: Example of Alternative Reactor Staffing Plan

Single and Dual Unit Plant Staffing

	Single Unit Station	Dual Unit Station
Plant Management		
Superintendent*	1	1
Assistant*	1	1
Operations		
Operations Supervisors*	1	2
Shift Supervisors*	6	6
Lead Operators/Foremen*	—	5
Control Operators**	11	16
Auxiliary Operators	11	16
Lead Fuel Handlers/Foremen***	—	3
Fuel Handlers	—	6
Technical		
Technical Supervisor	1	1
Professionals	6	9
Technicians	9	16
Maintenance		
Maintenance Supervisors	1	2
Craft and Repairmen	18	28
Security	11	16
Totals	77	128

*Senior Licensed Operator Qualifications
**Licensed Operator Qualifications
***Special Senior Licensed Operator Qualifications

Table 4.2:Personnel Requirements for Alternative Reactor Staffing Plan (Figure 4.3)

AN EXAMPLE OF OPERATOR COSTS

Basic Salary		
$8.00/hour x 2,080 hours/year	=	$16,640
Payroll Burden		
Federal Old Age Survivors Insurance		
Workmen's Compensation		
Pensions		
Life Insurance		
Company Contribution to the Thrift Plan		
38% of $16,640	=	6,323
Replacement Costs for:		
Two-week vacation		
Eight-day sick leave		
Nine-day holidays @1.5 rates		
14% of $16,640	=	2,330
		$25,293

Figure 4-4

The 30-year present worth factor at 8 percent interest per year is 11.26. Therefore, the present worth value of the 30 years of annual fixation costs would be 11.26 x \$53,000 ≅ \$597,000.

4.1.2 Fixed bed adsorption for C-14 control

This alternative has been proposed as a potential CO_2 removal system which removes all but 3 ppm of the CO_2 in the gas stream.$^{(20)}$

CO_2 is removed by passing the gas from the H_2O removal sieves through two 13-x sodium zeolite beds at approximately 5 atmosphere at 95°F.

Regeneration is accomplished with a smaller gas flow to maintain high CO_2 concentration in the stream. The gas is then diverted to the bottom of the CO_2 fixation tower where the gas bubbles through a saturated solution of $Ca(OH)_2$ which reacts with the CO_2 to form $CaCO_3$. The gas is next vented to the hood over the vacuum drum filter. The liquid and gas pass through the filter media to a gas-liquid separator.

The fabricated costs for the subsystem comprising H_2O regenerative beds and CO_2 regenerative beds are \$70,000 from vendor estimates. This compares with the costs given in reference 5.

The capital costs, based on the module factor of 7.5, are \$525,000 for the fixed bed adsorption process, including instrumentation, piping and valves.

Table 4-3

Equipment List for Fixed Bed Adsorption of CO_2

1. H_2O Sieves (3)
2. CO_2 Sieves (3)
3. Air Dryer
4. Air Compressor
5. CO_2 Fixation Tower
6. Vacuum Filter
7. Screw Conveyor
8. Dryer
9. Blower
10. Heater
11. Gas Liquid Separator
12. Lime Slaking Tank

The tanks, tower, filter housing, pump, piping and valves are made of 304 stainless steel.

The capital costs for the slaked lime subsystem can be obtained by scaling down the costs given in reference 15 of Chapter 3, to a lower limit of \$60,000. Thus, a lower limit for the total capital costs is \$585,000.

The annual fixation charges include:

Labor
\$25,000 x 1.0 = \$ 25,000
Maintenance and Replacement
\$585,000 x 0.07 = \$ 37,000
Chemicals = \$ 1,000
Fixed charges: 585,000 x 0.2 117,000
Total annual costs \$180,000

The 30-year present worth is: 11.26 x 180,000 = \$2,027,000

4.1.3 Fixed bed adsorption with caustic scrubbing for C_{14} Control

From vendor estimates,(16) the CO_2 can be concentrated by the fixed bed adsorption and regeneration process so that a gas stream of 10 cfm, one quarter of the original stream, is diverted to the caustic scrubber system.

As in the previous cost analysis, the capital cost for the fixed bed adsorption process is \$585,000.

The 10 cfm caustic scrubbing capital costs are scaled down from those for the 40 scfm case.

$$\left(\frac{10}{40}\right)^{0.6} \times \$100,000 = \$43,500$$

The total capital costs are: \$629,000. The annual fixation charges include:

Labor
\$25,000 x 1 = \$25,000

Maintenance and replacement	
$629,000 x 0.07	= $ 44,000
Chemicals	1,000
Fixed charges	
$629,000 x 0.2	$126,000
Total annual costs	$196,000

30 year present worth $196,000 x 11.26 = $2,207,000

We can see that caustic scrubbing is the most cost-effective C-14 control system.

In the following sections, only the caustic scrubber alternative will be considered for the PWR and LWR fuel reprocessing plant.

4.2 PWR off-gas C-14 treatment system

In pressurized water reactors, with the present limited data, it is estimated approximately 76 percent of the carbon-14 comes from the gas collection header and is held up in the waste gas decay tanks before being vented to the atmosphere. Since C-14 at the header may be predominantly in the form of hydrocarbons, it must be converted to CO_2 before it can be removed by scrubbing. This can be achieved by installing a recombiner upstream of the waste gas decay tanks.

Once decay of short-lived isotopes has taken place, the waste-gases must be processed. The same type of scrubber system that is described in the previous section for BWR off-gases provides adequate operational conditions for treating gas from the waste gas decay tanks. Note that control system operation for a PWR is intermittent, due to the fact that short-lived isotopes must decay in the waste gas decay tank before its contents are treated, so the scrubbing system will not operate continuously.

Capital and annual costs for a PWR off-gas removal system for C-14

The difference in cost between the BWR C-14 removal system and the PWR C-14 removal system is in the inclusion of the PWR recombiner. The recombiner is sized to accept the maximum flow from the PWR primary off-gas stream. This is approximately one cubic foot per minute. The purpose of the recombiner is to oxidize hydrogen to water. The carbon-14 control system takes advantage of this system to oxidize hydrocarbons to CO_2 and water. There is no other commercially available process unit that can perform this oxidization step in a manner that satisfies regulatory constraints.

A PWR recombiner system costs \$160,000 and includes a preheater, recombiner, post recombiner condenser and dryer, valves and control panels.(14) Without valves, piping and instruments, the fabricated cost is \$100,000 based on the cost module.

As indicated, the equipment design for the PWR system is essentially the same as for the BWR. The major differences are the intermittent operation of the PWR scrubbing system, an off-gas flow rate 40 times smaller, and inclusion of a recombiner.

A total of four cases were estimated for the BWR system (40cfm off-gas). The fabricated equipment costs for the four cases ranged from $9,000 to $19,000, with an average of $14,000. The addition of a $114,000 recombiner for the PWR system overshadows these fabricated equipment costs.

Therefore, one cost of $114,000 has been selected to represent the PWR case. This is the sum of $100,000 for the recombiner and $14,000 for the other fabricated equipment costs.

As indicated for the BWR, the fabricated equipment cost represents 13.3 percent of the total capital cost. Therefore, the total capital cost of the PWR is estimated at $855,000.

Annual costs for the system are estimated by the same method used for the BWR system. The labor costs assume one additional staff member at the qualification level of an operator at $25,000 per man year.

Labor	
$25,000 x 1.0 =	$ 25,000
Maintenance and Replacement	
$855,000 x 0.07	$ 60,000
Chemicals	1,000
Fixed charges	$171,000
Total annual costs of fixing CO_2 as $CaCO_3$	$257,000

The 30-year present worth factor at 8 percent interest per year is 11.26. Therefore, the present worth value of the 30 years of operating costs would be 11.26 x \$257,000 ≅ = \$2,894,000.

Retro-fit costs and schedules

It is impossible to provide an accurate estimate of either a generic cost or generic schedule of system retro-fits. All retro-fits are site and facility specific and will surely vary considerably. The important factors that affect cost and schedules include:

- Facility operating schedules
- Available space envelopes for the installation of new equipment
- Accessibility of required tie-in points for both process functions and utility support systems.
- Availability of space envelopes for control and support functions
- Extent of licensing activities.

As an example of what would be involved in retrofitting a caustic scrubbing unit for the removal of C-14, a sample case is provided. The sample case chosen is the Davis-Besse Nuclear Power Station, Unit I, which is a PWR.

Figure 4.5 shows the location of the waste gas decay tanks. They are housed at elevation 545' of the auxiliary building, elevation 585' being ground level. The room which houses the waste gas decay tanks does not have sufficient room to house a scrubbing system since the tanks occupy most of the space in the room.

Figure 4-6 shows the waste solidification and drumming room. Note that it is diagonally opposite the waste gas decay tank room and at an elevation of 585'. This room also does not have sufficient space to house the C-14 scrubbing system.

Since the waste gas decay tank room is below ground level, it would be extremely difficult to build a room adjacent to it. The other alternative is to construct a room alongside the auxiliary building adjoining the waste solidification and drumming area. This would require new construction and knocking out a portion of the auxiliary building wall to allow entrance to the scrubber room. The arrangement is shown in Figure 4-6.

A gas line would be connected to each waste gas decay tank, brought to a common junction and run vertically upward from the waste gas decay tank room to the auxiliary building ceiling, then down the side of the building to the drumming area and finally into the scrubber room. This path represents the least number of problems for retrofitting as far as knocking holes in walls, and avoiding existing equipment. Holes would have to be cut in floors to run the lines vertically. The top of the auxiliary building is open and presents no barriers.

Table 4-4 serves to illustrate the major items that must be included in the retrofit of the C-14 cleanup system and an estimate, where applicable, of the features and requirements of each item.

It is seen that each reactor layout has its own specific problems. The unusual construction problems are those encountered in working in an actual or potentially contaminated area. In addition special security procedures come into place when working in controlled access areas.

Preliminary estimates associated with the Davis-Besse reactor would suggest the additional structure and other modifications

TABLE -4

Retrofit Items	Size	Amount	Materials	Special Requirements
Alterations to structures				
1. Addition of room to house caustic scrubbing system	20'x20'x30' (2' wall thickness)	1	concrete rebar insulation paint	Seismic Category I
2. Penetrations	4"	35-100		
Utilities Support				
1. Water				
2. Chemicals				
3. Electrical				IEEE Std. 279 (protection system) IEEE 300 (criteria for class IE) IEEE 323 (Qual. of class I eqpt.) IEEE 336 (N45.2.4) (Q. A. req'ts) IEEE 344 (seismic Qual.)
4. Steam				
5. HVAC				
6. Decontamination System (Including Floor drains)				
Control and Instrumentation				
1. Instrument Leads				(see above IEEE Criteria)
2. Motor Control Centers				
Flow Devices				
1. Piping	2"	500'	Stainless Steel	ASME class 1
2. Valves (with controls)	2"	10	Stainless Steel	ASME class 3
3. Connectors (elbows, tees, etc.)	2"	50	Stainless Steel	ASME class 3
Supports				
1. Hangers		50		

Descriptions of Representative Retrofit Items

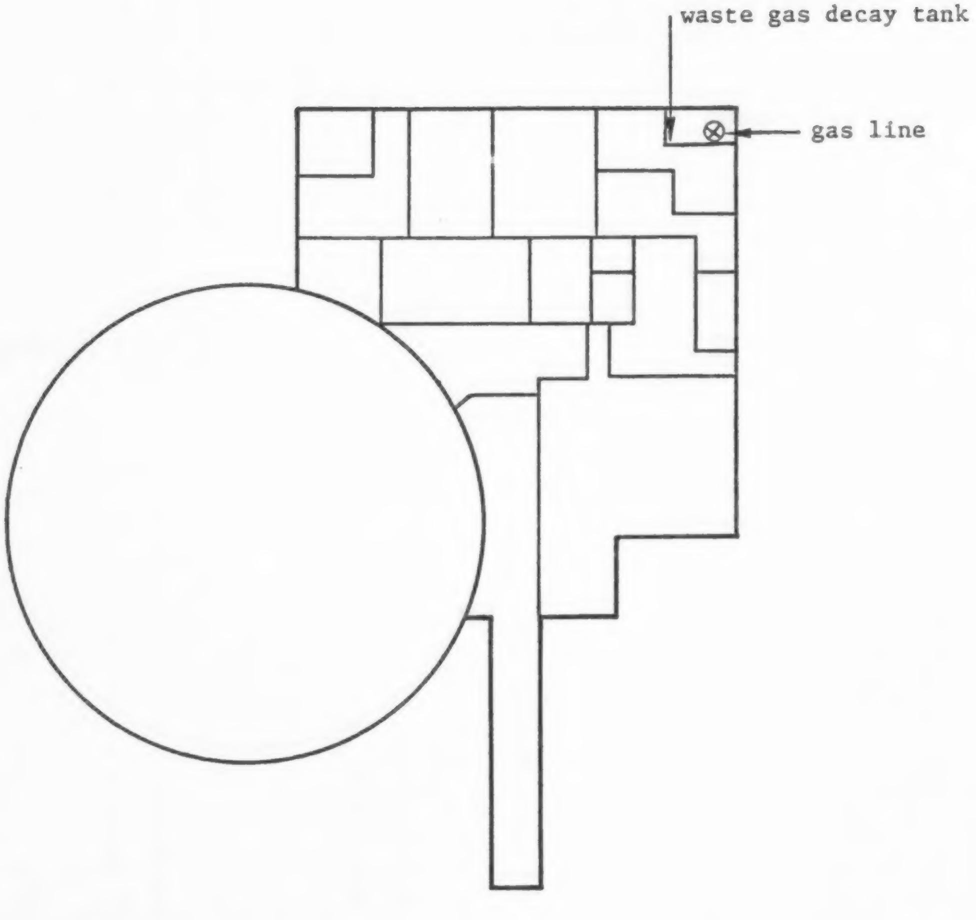

waste gas decay tank
gas line

Figure 4.5: Davis Besse Nuclear Power Station Elevation 545

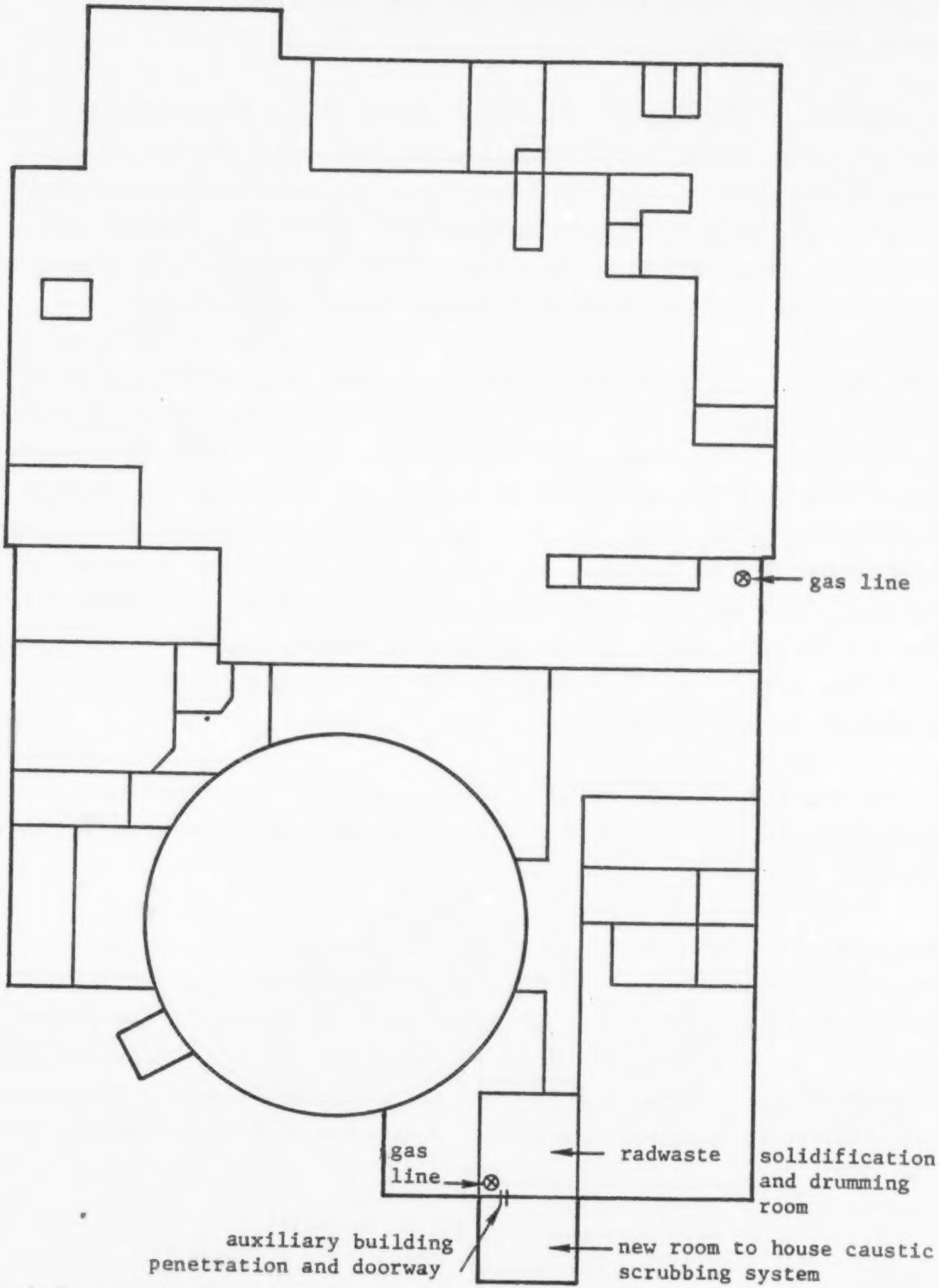

gas line
gas
line
radwaste
solidification
and drumming
room
auxiliary building
penetration and doorway
new room to house caustic
scrubbing system

Figure 4.6: Davis Besse Nuclear Power Station Elevation 585

might approach $500,000 while the time requirement may be about six-months.

It should be emphasized that each retrofit is site-specific and the particular retrofit analyzed here only serves to illustrate some of the considerations in retrofitting the C-14 cleanup system. Extensive engineering work, well beyond the actual or intended scope of work, would be required to complete the engineering and cost estimate on even one reactor.

The largest cost item associated with the retro-fit of a C-14 control system for any LWR will be the production revenue lost during the final phase of construction, tie-in, and commissioning. The lost revenue from an outage of a reactor can range from $300,000 to $500,000 per day. If the system is constructed independently and tied-in to the off-gas system at a scheduled outage, the costs in lost revenue would be moderate. Depending on how the construction of a C-14 control system interferes with the operation of a reactor the outage might range from one week to one month.

Licensing activities include preparation of documents and participation in hearings. These may require extensive outlays of time and money.

The retro-fit of a system into a radioactive facility entails unusual, and additional, construction problems. Workers must be trained in radiation protection and control procedures. Construction barriers must be established to control loose contamination. Finally, special security systems and procedures are needed to prevent industrial sabotage of a radioactive facility.

Table 4-5 summarizes the cost factors directly related to retrofit. The costs are presented as ranges.

The schedule for the installation of a retrofit carbon-14 control system could range from 3 to 5 years. The major activities required for retrofitting include:

Initial analysis in support of system choices
Engineering
Procurement of equipment
Construction of the system
Checkout and startup
Licensing

A possible schedule is shown in Figure 4.7.

As indicated on page 5-4, for a newly designed LWR there are no engineering, procurement, or construction problems that would represent a change in the basic facility schedule. All of the equipment required for this system is commercially available.

Table 4-5

COST FACTORS DIRECTLY RELATED TO RETROFIT

	Range $ x 10^6
Lost production revenue ranging from 1 week to 1 month	2 to 15
Licensing activity costs	0.1 to 1.0
Unusual construction problems	0.05 to 0.5

Typical Retrofit Schedule
For a C-14 Control System

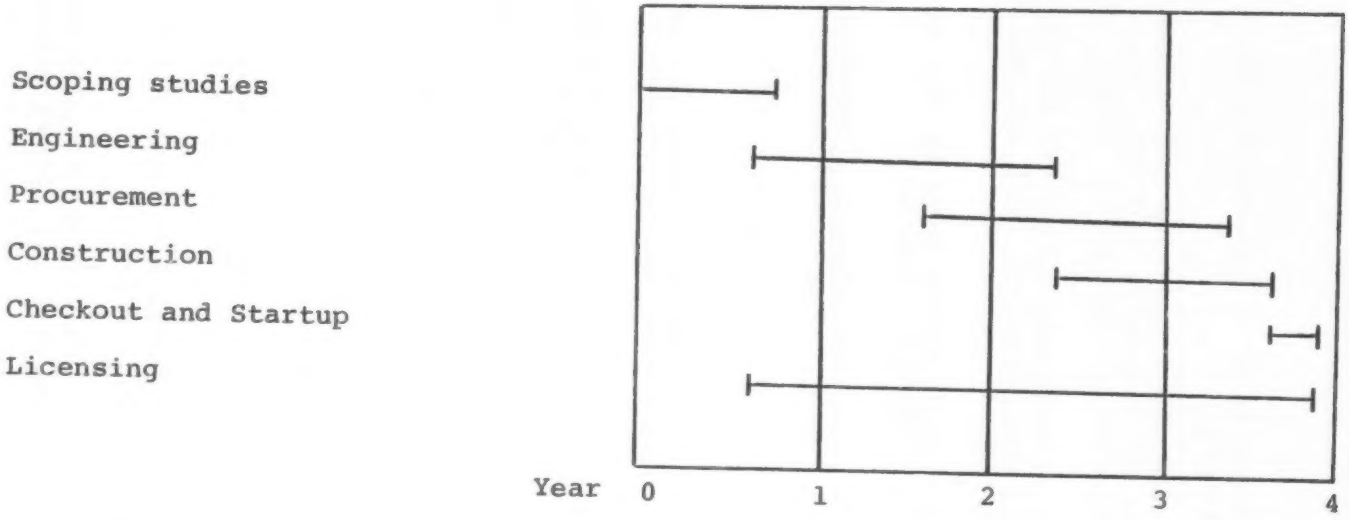


Figure 4-7

Chapter 4 References

1. Private communication between SAI and the Nuclear Regulatory Commission.

2. "Environmental Radiation Protection for Nuclear Power Operations", Proposed Standards (40 CFR 190), Supplementary Information, January 5, 1976.

3. Private telephone communication between SAI and Bechtel Power Corporation, Gaithersburg, Maryland and San Francisco, California.

4. Private telephone communication between SAI and Elwood Nuclear Safety Inc., Buffalo, New York.

5. E. D. North and R. L. Booth, Fission Product Gas Retention Study. Final Report, ORNL-TM-4409, 1973, pp. 51-52

6. Private telephone communication between SAI and Norton Co., Akron, Ohio.

7. Private telephone communication between SAI and ACS Industries, Inc., Woonsocket, Rhode Island.

8. Private telephone communication between SAI and Spray Engineering Co., Burlington, Massachusetts.

9. Private telephone communication between SAI and Chemineer Agitators, Inc., Dayton, Ohio.

10. Private telephone communication between SAI and Duriron Co., Inc. Dayton, Ohio.

11. Private telephone communication between SAI and Armco Steel Corp., Baltimore, Maryland.

12. J. Happel and D. G. Jordan, Chemical Process Economics, Marcel Dekker, Inc., New York, 1975, p. 235.

13. Private telephone communication between SAI and Vacco Industries, El Marte, California.

14. Private telephone communication between SAI and WACO Associates, Inc., Lafayette Hill, Pennsylvania.

15. Private telephone communication between SAI and a chemical supply warehouse.

16. Private telephone communication between SAI and Cosmodyne, Inc., Torrance, California.

17. I. Leibson and C. A. Trischman, "Spotlight on Operating Cost", Chemical Engineering, May 31, 1971, pages 69-74.

18. Portland General Electric Company, Docket No-50-344, Operating License and Appendices

19. WASH-1130 Revised, Utility Staffing and Training for Nuclear Power, June 1973.

20. R. A. Brown, Reference Facility Description for Volatile Radioisotopes, Idaho National Engineering Lab., 1977 (in publication).

CHAPTER 5. DESIGN MODIFICATION TO NEW LWR FACILITIES FOR THE CONTROL OF C-14

Design changes to new light water reactors that might be implemented in order to remove carbon-14 from the facility effluents may be significantly different from the retrofit designs of Chapter 4. Following sections discuss design changes for boiling water and pressurized water reactors based on criteria this study has identified or assumed in Chapter 2 (Tables 2-2 and 2-3) regarding the major carbon-14 release pathways.

It should be pointed out that definitions of these pathways are preliminary in nature and based on a limited number of carbon-14 measurements in reactors of earlier design. A review of Chapter 2 indicates that greater than 99 percent of the C-14 discharged from a BWR is reported to be from the main condenser air ejector exhaust. But in the case of a PWR, approximately 16 percent of the C-14 is reported to be discharged from the fuel handling building ventilation and approximately 12 percent is said to be discharged in containment purges. The calculated value for the containment purge discharge using standard leak rates is less than about 0.01 percent. In contrast, reported values of C-14 discharges from both the PWR fuel handling building and the containment are based on laboratory data which approach the lower detection limit of the analytical methods utilized.

5.1 BWR design modifications

The only source for carbon-14 in a BWR plant considered by this study, because it contains 99 percent of the off-gas carbon-14, is the exhaust from the SJAE condenser vent. Following the condenser, a recombiner can be installed which would eliminate radiolytic H_2 and O_2 and cool the gas stream to about 140°F. Gas flowrate at this point should be no more than 40 cfm. This stream could be treated quite readily in a caustic scrubber following the design principles of Chapter 4. It is emphasized that the effluent gas from the recombiner need not be dried if its immediate destination is a caustic scrubbing column. New plants incorporating both systems may wish, therefore, to leave out a drying step after the recombiner. It is currently thought that a recombiner system will be required in new BWR facilities. The resultant cost of such treatment would be essentially the same as reported in Chapter 4.

5.2 PWR Design Modifications

Carbon-14 isolation from PWR off-gas streams will be more involved than in the BWR, especially if the reported pathway proportions are confirmed by future reactor sampling programs. Sources subject to potential design modification would include the gaseous radwaste treatment system, secondary system condenser air ejector, fuel handling building ventilation, and containment building purge exhaust. Present projections of C-14 discharges rates would suggest that the treatment of these four streams may result in the capture of greater than 99 percent of the C-14 presently being released.

The condenser air ejector flow of 25 cfm could be treated by a scrubber system similar to the one discussed in Chapter 4. It may join tasks with the system proposed for the primary gaseous radwaste stream.

The fuel handling building ventilation flow of 2470 cfm presents a different type of engineering problem. It is a relatively large flow with a very low concentration of C-14. Instead of treating the large flow of air, internal recycle of 95 percent of the ventilation air would reduce the amount of gas to be treated to 120 cfm. This volume of gas could more easily be treated by a caustic scrubber system.

The containment building purge represents the most challenging problem because the flow is both large and intermittent. The volume to be treated is approximately 10^6 cubic feet four times a year. The treatment of a very large flow of air for recovery low concentrations of carbon dioxide is presently not a commercial practice. A great deal of work is required in both engineering and development aspects to resolve this problem. One potential solution would be to store the purge gas and treat it on a continuous, but diminished, rate prior to the next purge.

5.3 Reduction of carbon-14 by control of parent elements

Table 2-1 indicates the amount of carbon-14 produced from various parent elements. It has been suggested recently that some carbon-14 control may be effected prior to its creation by limiting the amount of parent substances subject to irradiation.(1) Such control would reduce releases from LWRs and separations facilities.

Two neutron activation reactions were shown to dominate the production of carbon-14:

$^{17}O\ (n, \alpha)\ ^{14}C$ and $^{14}N\ (n, \rho)\ ^{14}C$. In fuel, the amount of oxygen present is completely dependent on the mass of fuel required and is therefore somewhat inflexible to adjustment. The oxygen reaction accounts for approximately 20% of the fuel source term, as long as assumptions about the ^{14}N reaction are realistic. The amount of ^{14}C resulting from ^{14}N is dependent on an educated assumption as to the extent of nitrogen impurities in fresh fuel. A decision regarding nitrogen control depends on the source of ^{14}N in the fuel. If nitrogen is present in raw materials, control may be impossible. If the greatest portion is introduced during fuel manufacture, control may be achieved, though the final level of prevention depends on a quantitative knowledge of nitrogen impurities in the fuel. According to the data in Table 2-1, about 80% of the fuel source term would be subject to reduction by control of nitrogen impurities. The greatest effect of a diminished fuel source term would be felt at the separations stage, though reactor effluent levels might also be reduced.

The amount of coolant oxygen is fixed, and the amount of coolant nitrogen is known with little confidence. As stated in

Section 2.1.2, the source term for coolant carbon-14 resulting from neutron activation of ^{14}N is assigned a nominal value awaiting measured levels. In no case at present could carbon-14 be considered to be subject to control by elimination of parent elements in the coolant.

Chapter 5 References

1. Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF-76-0701, July 11-16, 1976, Denver, Colorado. (page 381).

CHAPTER 6. CURRENT SPENT NUCLEAR FUEL REPROCESSING EFFLUENT TREATMENT SYSTEMS

Spent nuclear fuel contains significant quantities of fissile materials. This fissile material includes both U-235 from the original enriched uranium fuel and plutonium generated by reactor operations. It represents a potentially large source of energy if the fissile material is recycled back to reactors as recycled fuel. The first step in the recycle of this fissile material is the chemical reprocessing of spent nuclear fuel. Figure 6-1 is a block flow diagram showing the major process units of such a reprocessing facility. These operations generate process off-gases, process liquid waste, and liquid effluents.

Some difficulty arises at this time in treating reprocessing facility off-gas systems in a generic fashion. There is only one such facility of the modern type where designs have been finalized and constructed. That facility is the Barnwell Nuclear Fuel Plant (BNFP) and it is here used to exemplify current concepts in process design. Newer plants, such as that proposed by Exxon Nuclear Company, may be forthcoming. An analysis such as this one, however, requires concrete information available only from current experience and criteria. Specific data in this analysis come from the BNFP Final Safety Analysis Report for the Separations Facility.(1) Other sources discuss details of the Separations Facility as well.(3,4)

NUCLEAR FUEL REPROCESSING FLOW DIAGRAM

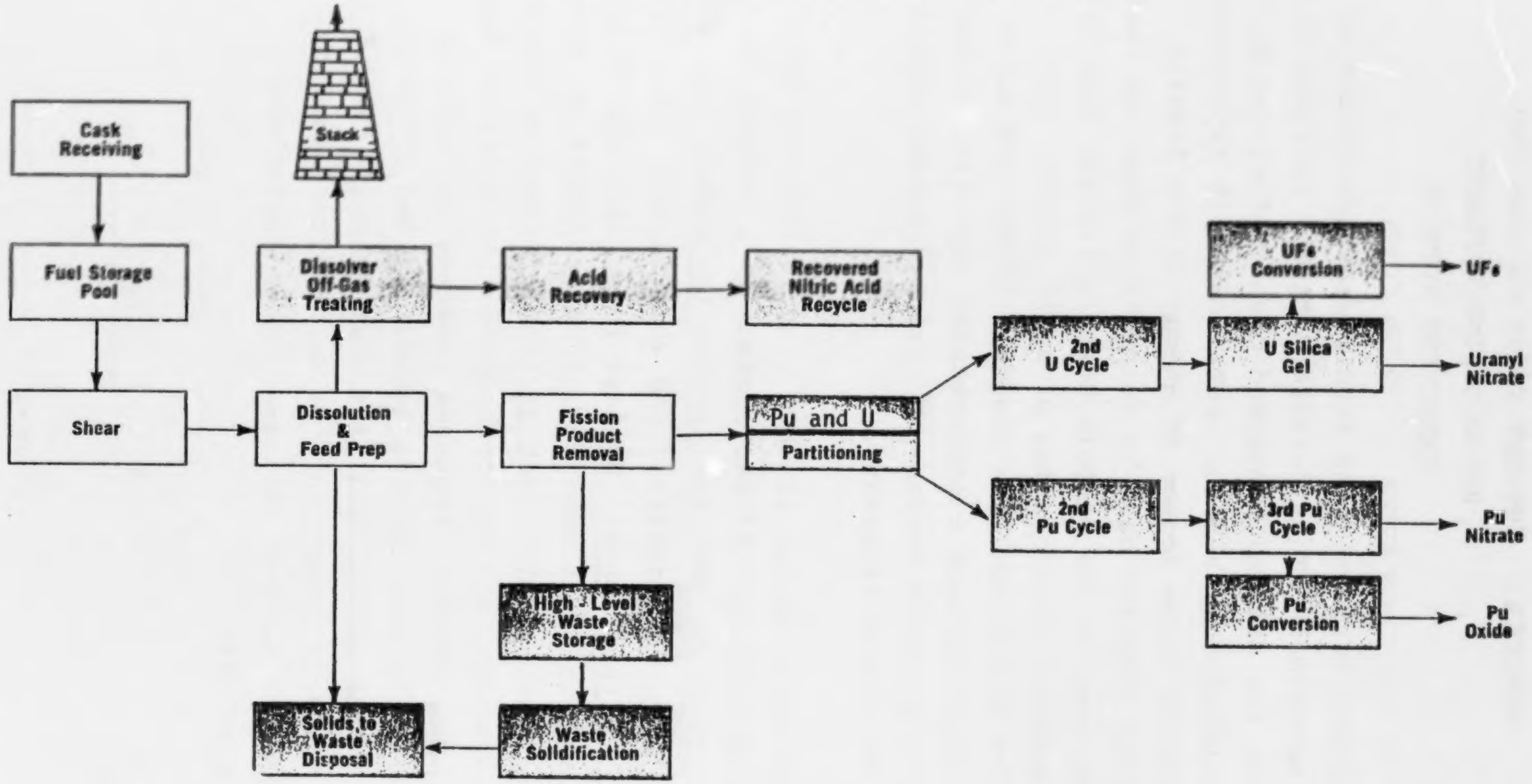

Cask Receiving
Fuel Storage Pool
Shear
Stack
Dissolver Off-Gas Treating
Dissolution & Feed Prep
Solids to Waste Disposal
Acid Recovery
Recovered Nitric Acid Recycle
Fission Product Removal
High - Level Waste Storage
Waste Solidification
Pu and U Partitioning
2nd U Cycle
2nd Pu Cycle
UF₆ Conversion
U Silica Gel
3rd Pu Cycle
Pu Conversion
UF₆
Uranyl Nitrate
Pu Nitrate
Pu Oxide

Figure 6-1

The nuclear fuel reprocessing flow diagram of BNFP is shown in Figure 6-1. Treatment systems are in various stages of development. The cask receiving and fuel storage pool areas are primarily subject to contamination via leaking fuel rods which spill gaseous contents into the cooling water. Some of these gases partition into the building atmosphere, though measurements are not available for BNFP. Presently, this atmosphere is purged on a continuous basis through a filtration system. The cooling water is continuously demineralized and a certain fraction of leaked nuclides are removed, while the rest presumably remain in the water. They do not, however, escape to the environment in an uncontrolled way. After the fuel storage pool, fuel travels to the shear area for hull removal. Process effluents from this point forth are separately treated and are discussed in the following chapter.

6.1 Process off-gas treatment

A process off-gas treatment system is part of the basic flowsheet of all nuclear fuel reprocessing facilities. Examination of Figure 6-1 reveals several different needs for off-gas treatment, not all of which pertain to radiological hazard directly. The general treatment processes involve:

- removal of process condensates derived from off-gas condensers
- iodine (predominantly longer-lived ^{129}I) removal by both wet chemical scrubbing and fixed bed adsorption

- removal of nitric acid and nitric acid fumes ("NO_x's") formed from thermal and radiolytic decomposition of dissolved nitric acid and nitrates
- final high efficiency particulate filtration

A schematic diagram of the interrelations of these treatment processes is provided in Figure 6-2. More will be said of the specific processes at BNFP in Chapter 7. However, the two major process sections are described here for reference. The first section handles and treats the off-gas from dissolvers and is referred to as the dissolver off-gas system (DOGS). The off-gas from the majority of the process vessels enters the treatment system following the NO_2 Absorber Eductor. At this point, the off-gas treatment system is referred to as the vessel off-gas system (VOGS). The flow of gas in the dissolver off-gas system can be expected to range from 300 to 500 scfm, while the flow in the vessel off-gas system can range as high as 6,000 CFM. The combined DOG and VOG streams are treated further, filtered and mixed with air from the ventilation system for release through the Acid Fractionator Overhead Vaporizer and the 100-meter main stack.

Other harmful materials besides carbon-14 are under consideration for removal from the waste gas stream such as fission product tritium and krypton-85. Steps to remove one off-gas constituent usually relate to others; such relations will be indicated where observed.

Reprocessing Facility Off-Gas
Treatment Flow Diagram

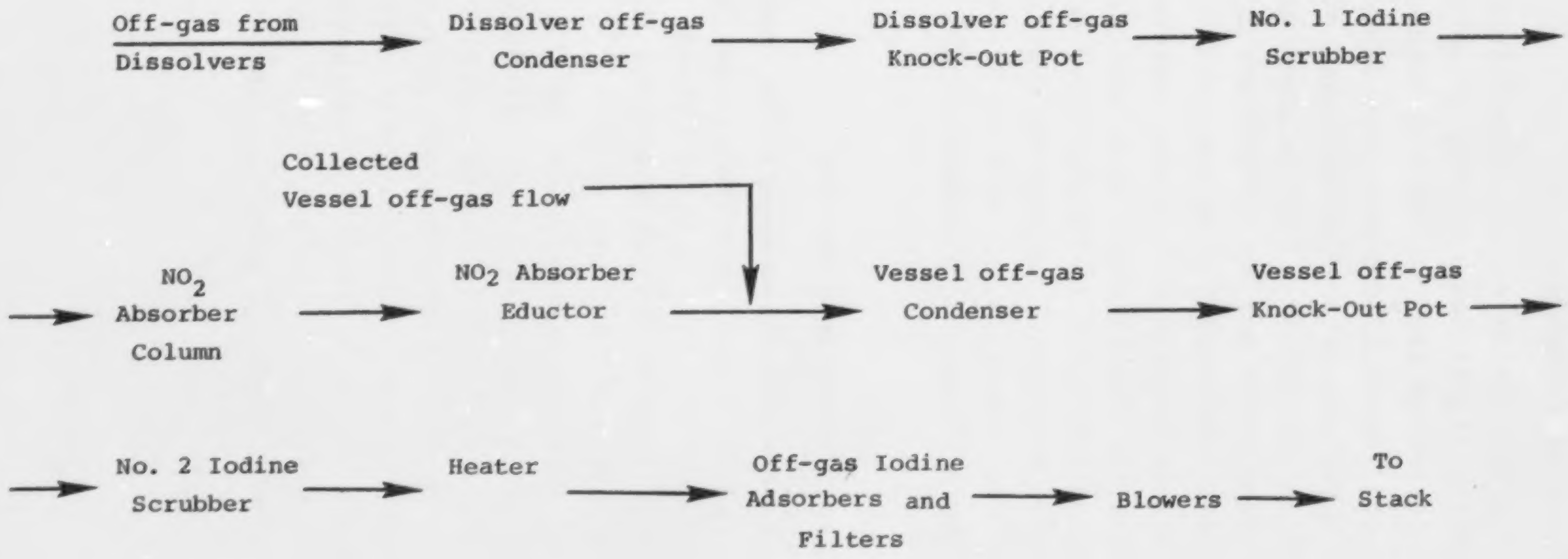

Off-gas from Dissolvers
Dissolver off-gas Condenser
Dissolver off-gas Knock-Out Pot
No. 1 Iodine Scrubber
Collected Vessel off-gas flow
NO_2 Absorber Column
NO_2 Absorber Eductor
Vessel off-gas Condenser
Vessel off-gas Knock-Out Pot
No. 2 Iodine Scrubber
Heater
Off-gas Iodine Adsorbers and Filters
Blowers
To Stack

Figure 6-2

6.1.1 Process off-gas condensation

Gas streams from both the dissolver and composite vessel off-gas systems contain significant amounts of water vapor. This is condensed in the DOG and VOG condensers, allowing the dried gas stream to continue and condensate to be removed in a knock-out pot. The condenser also provides a decontamination factor. Particulates may be trapped on the wet tube faces and be carried off in the condensate. In normal conditions, a DF for particulates of 100 or greater may be available from the condenser alone, though this varies considerably with operating conditions and type of activity; it is therefore not claimed for regulatory purposes. Condensers are not seen as a removal system for carbon-14, as most of it is gaseous after exposure to the oxidizing environment of the dissolver.

6.1.2 Removal of radioiodine from off-gas

There are two systems specifically designed to remove residual, fission product iodine from the off-gas stream at the BNFP Separations Facility. A third method, called the Iodox process, has been studied at Oak Ridge National Laboratory(5,6) and considered for use at BNFP.(4) It is currently considered unsatisfactory for use at the reprocessing plant, though it is a potential control technology. Finally, after careful review of iodine pathways, a small fraction of entering radioiodine is expected to volatilize after off-gas treatment steps in the pre-stack water vaporizer, a potential limitation to the overall decontamination factor (DF). Solutions to ameliorate this condition are proposed(4) but generally are modifications of present techniques so will not be separately considered.

6.1.2.1 Wet chemical scrubbing

This method employs a scrubbing column and a scrubbing agent called mercuric nitrate. Both elemental and organic iodine are complexed by mercuric ion in this solution approximately 8 M in nitric acid At BNFP, the DOG stream and certain non-condensible compounds enter the #1 Scrubber and pass upward through a solution 0.2 M in mercuric nitrate and 6-8 M in nitric acid. The exiting stream passes through the NO_2 Scrubber and the NO_2 Absorber Eductor and to be combined with the very high volume VOG stream. This stream passes through the #2 Scrubber where iodine is again removed by reaction with mercuric nitrate in nitric acid. Each column is expected to have an iodine DF of 10.(4)

Scrubbing solution is initially charged via the #2 Scrubber. The bottoms from #1 Scrubber are drained to the Intermediate Level Liquid Waste system periodically, and it is charged from the bottoms of #2 Scrubber. The second column is then freshly charged.

This system has only speculative value for carbon dioxide removal (recall that most carbon will be oxidized in the dissolver, if not subsequently). It is thought that the scrubbing solution is so heavily acidic that it will largely prevent dissolution of CO_2, and consequently most of the gas will pass out of the system.

6.1.2.2 Dry fixed-bed adsorption

The out-flow of #2 Iodine Scrubber passes a roughing and a fine filter, then a dry fixed-bed adsorber packed with silver zeolite (AgZe). AgZe is formed by exchanging sodium on zeolite with silver using silver nitrate. It will react with either elemental or organic iodine. A fully exchanged AgZe filter will retain 0.09 grams of iodine per cubic centimeter of bed up to 1 percent breakthrough. The iodine DF is at least 100.(4) Combined with the scrubber, an overall DF for iodine in the Process Off-Gas System of 10^4 is effected, surpassing the suggested factor of 10^3 (7) for 150 day cooled LWR fuel.

This filter is not seen as a carbon-14 removal system by itself, though zeolite is known to act as a molecular sieve for CO_2. Physical hold-up may therefore occur, but this system is used without a purge system when treating iodine. Thus, it cannot of itself trap and retain $^{14}CO_2$.

6.1.2.3 Iodox process

An advanced process under development at Oak Ridge National Laboratories is called the Iodox process. It has not been accepted for use at BNFP or elsewhere, but experimental results are available and indicate this is a highly effective system for iodine removal.(5,6) The off-gas stream is placed in contact with 16-20 M nitric acid in a bubble-cap scrubbing column, possibly similar to the one used presently at BNFP for NO_2 absorption. Both organic and elemental iodine are precipitated as iodic acid which is collected in the bottom liquids. Experiments in a 6-stage fractionating column indicate decontamination factors ranging from 30 in 17 M nitric acid to 9×10^5 in 20 M nitric acid. Such high decontamination factors may be required, especially if more highly burned LWR fuel, mixed oxide fuel or LMFBR fuel are to be processed in a separations facility, or possibly fuel which has undergone a shorter cooling period.

It is difficult to predict the usefulness of this process with respect to carbon-14 removal. Lacking more specific information, this report merely indicates the system for a potential off-gas treatment, to be considered at the appropriate time, in keeping with our efforts to analyze C-14 systems as one part of an integrated off-gas treatment process.

6.1.3 Removal of nitrogen oxides by wet scrubbing

Nitrogen oxides, such as N_2O, NO, NO_2, N_2O_5 or, more generally, NO_x, are released to the cover space of the spent fuel dissolver due to thermal, chemical and radiolytic decomposition of nitric acid and nitrates.

Nitrogen dioxide (NO_2) is scrubbed from the off-gas at BNFP by passing the gas upwards through a multi-stage bubble cap column with water as the scrubbing agent. Cooling coils remove heat generated by the exothermic absorption reactions. NO partially reacts with oxygen to form NO_2, which is absorbed with other NO_2 as nitric acid. NO_2 is reduced by a factor of about 10 and NO by a factor of 4; approximately 31 pounds per hour of NO_2 plus NO and 21 pounds per hour of N_2O are discharged.(1) This process does not significantly reduce carbon-14, assumed to be present as $^{14}CO_2$, by water dissolution, probably because the water becomes rapidly acidic with the H_2O - NO_2 decomposition reaction. Special consideration will have to be given to removal of N_2O, as well as CO_2, in the event cryogenic extraction of noble gases is required.

6.1.4 Cryogenic distillation for noble off-gas treatment

This concept is discussed in a general manner here, for it is only a proposed control technology for noble gas separation at a commercial facility. It has been used effectively at smaller facilities such as the Idaho Chemical Process Plant;(2) this experience is considered in reports on applicability of cryogenic distillation at BNFP.(4) It presently appears that another method, fluorocarbon absorption, is favored because it is less sensitive to off-gas impurities that foul a cryogenic system.

6.1.4.1 Removal of noble gases by cryogenic distillation

This technique capitalizes upon differences in boiling point of the several off-gas constituents. At temperatures achieved with a liquid nitrogen coolant, the various gases fractionate as follows:

Liquid, Solid or Slush	Gas
Xe (B.P. = -107°C)	N_2 (B.P. = -196°C)
Kr (B.P. = -153°C)	O_2 (B.P. = -183°C)
CO_2 (B.P. = -79°C)	CO (B.P. = -190°C)
	H_2 (B.P. = -253°C)

(All boiling points are for one atmosphere cover pressure)

Some constituents of the gas stream are liquified, fractionally distilled and the distillates collected. Typical purity limitation is encountered due to vapor pressures of the liquid components,* impurities in the reactor off-gases from which the liquor is composed and radiolytic products formed in the distillation apparatus. Nevertheless, product gases become separated to a high degree and they can be stored at a greatly reduced volume and subsequently released following decay holdup. They might also be bottled and stored for long-term decay. The latter alternative provides for nearly zero release of noble gas, while both processes effectively remove iodine.

The distillation apparatus are similar in each system reviewed. Feed gas is deoxygenated to prevent excessive ozone formation. It is then dried and chilled in a regenerative pre-cooler (cold-trap) to remove most high boiling components which would clog the recovery column or reduce its efficiency.(3) Catalytic recombination over rhodium(2) or platinum-palladium surfaces can reduce elements which foul the cryogenic system, but a precooling stage is required prior to the distillation equipment as well. The gas stream then passes, in a countercurrent fashion, in contact with liquified nitrogen (LN_2) coolant in which all products except nitrogen, ozone, traces of oxygen and hydrogen, and carbon monoxide are condensed. The vent gases which now contain very little Kr-85, are passed through a regenerative precooler and vented to the atmosphere. The liquid fraction passes to a separation column where it is fractionally distilled and the vaporized constituents are removed and held separately. Alternatively, the liquified products may be left as a mixture and stored for decay in a holdup tank, then released after 45-90 days (the time period is only for illustration). If bottling is employed, on-site shielded remote handling facilities must be available for the Kr-85 fraction until it is packed in a transfer cask. A further design feature on some separation columns is a recycle line to the feed gas holdup tank for any occluded or entrained radioactive species.(3) The cryogenic apparatus is packaged in a positive pressure inert atmosphere to prevent leakage hazards.

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* Approximately 0.01 percent of the Kr and iodine and 0.025 percent of the Xe are vented with the carrier gases.

6.1.4.2 Application of cryogenic distillation to carbon dioxide removal

The extent that such a system may remove carbon-14 from the waste gas stream is undetermined, though it will depend very much on the specific apparatus and process conditions. Because the cryogenic system requires a recombiner system for oxygen, it is assumed that most carbon in the off-gas stream will be oxidized and thus in a gaseous state. The precooling step should effectively remove CO_2 from the stream to prevent system fouling, but the high-boiling compounds de-entrained at this step would be released. While cryogenic distillation is not itself judged to be a satisfactory control technology for C-14 for the reasons described on page 3-20, an alternative control system must be applied in advance of cryogenic noble gas equipment.

6.1.5 Selective absorption of gaseous radwastes into liquid dichlorodifluoromethane

A general discussion of fluorocarbon absorption as a gaseous radwaste treatment system and potential system descriptions for BWR's and PWR's were provided in Section 3.2.2. It was emphasized in Chapter 3 that this system cannot currently be considered a complete carbon-14 removal technology providing a stable final form. A general design for a process applicable to the BNFP Separations Facility was provided by Murbach, et al.(4) This design exemplifies a "current", feasible reprocessing off-gas system without, of course, specifying the nature of the final stable products.

A fluorocarbon absorption device would probably be placed in the stream following NO_2 treatment. The smallest flow-rate available, with the minimum level of fouling agents, is at that point. Pre-treatment for residual NO_2 and moisture, as well as CO_2, is suggested in the BNFP design to minimize system fouling. Pre-treatment is presently done with molecular sieves, though more extravagant systems may be required. As suggested in Chapter 3, allowing any but a small amount of $^{14}CO_2$ into the fluorocarbon system might create more problems than it solves.

However, firm understanding of these relationships is only possible utilizing data from a prototypical system.

The process as presently outlined involves pre-treatment, cooling, absorption, stripping and regeneration of the solvent in liquid form. Product forms are invariably gaseous, requiring physical or chemical stabilization, and the final form is dependent on isolation requirements. A system of this sort will most probably provide isolation of radioactive noble gases from a reprocessing plant,(4,8) while decisions regarding its efficacy for other nuclides await further study. Fluorocarbon absorption is not presently considered a carbon-14 isolation technology per se, though its use in an integrated system should surely be examined as design requirements and constraints become more specific.

6.2. Process liquid waste and liquid effluent

There are two radioactive liquid waste streams generated by the operation and maintenance of a nuclear fuel reprocessing facility. These two streams are referred to as high-level liquid waste (HLLW) and intermediate-level liquid waste (ILLW). They are collected, concentrated, stored for an interim period of time, and then solidified.

High-level liquid waste is defined as the aqueous raffinate from the first decontamination cycle of a nuclear fuel reprocessing facility. This waste stream contains greater than 99 percent of all the non-volatile fission products associated with incoming spent nuclear fuel.

The HLLW stream is generated at a rate of approximately 1,200 gallons per metric ton of uranium reprocessed. It is then concentrated to between 150 and 300 gallons per MTU. Following some interim storage period, the HLLW is solidified as a stable waste product form.

Intermediate-level liquid waste is composed of several aqueous wastes. These aqueous wastes include solvent wash wastes, laboratory wastes, floor drainage, and equipment and facility decontamination wastes. These streams are collected at one point and concentrated to a pre-determined concentration. After some period of interim storage as liquid, this waste is also solidified as a stable solid.

Several of the process unit operations of a nuclear fuel reprocessing facility generate process condensates. These operations include both the waste and product concentrators and the off-gas system condensers. After treatment, this material is released to the environment as an effluent. In the case of the Barnwell Nuclear Fuels Plant, treated process condensates are released to the main process stack as a vapor.

The treatment processes for liquids usually generate a terminal storage material and medium. At BNFP, the two liquid streams will be solidified, and the vaporized liquid condensates treated as off-gas prior to release. The latter treatment system was examined in Section 6.1. These alternatives for liquid waste provide isolation of nuclides, including carbon-14, from the environment. The method of process condensate vaporization allows for general decontamination, but nothing specific for carbon-14 as a gas. Application of the methods of Chapter 8 will mitigate this release.

Chapter 6 References

1. Allied-Gulf Nuclear Services. *Barnwell Nuclear Fuel Plant Separation Facility Final Safety Analysis Report*, Section 4, October, 1973.

2. Bendixsen, C. L. and G. F. Offutt. *Rare Gas Recovery Facility at the Idaho Chemical Processing Plant*. United States Atomic Energy Commission, IN-1221, April, 1969.

3. *Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle*. Volume II, Section 13.0, United States Energy Research and Development Administration, ERDA-76-43, May, 1976.

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5. Groenier, W. S. *An Engineering Evaluation of the Iodex Process: Removal of Iodine from Air Using a Nitric Acid Scrub in a Packed Column*. Oak Ridge National Laboratory, ORNL-TM-4125, February, 1973.

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8. Stephenson, M. J. and R. S. Eby. "Development of the FASTER Process for removing krypton-85, carbon-14 and other contaminants from the off-gas of fuel reprocessing plants". *Proceedings of the Fourteenth ERDA Air Cleaning Conference*, August, 1976, (in publication).

CHAPTER 7. PROJECTED CARBON-14 CONCENTRATIONS AND BEHAVIOR IN NUCLEAR FUEL REPROCESSING EFFLUENT TREATMENT SYSTEMS

This chapter will analyze the expected carbon-14 concentrations, chemical forms and behavior in the effluent systems of a nuclear fuel separations facility. This information is needed to exhibit parameters used in design of an effluent control system. Knowledge of the distribution and quantity of releases from the several pathways which carry carbon-14 to the environment allows the designer to choose cost-effective treatment principles and devices.

As in Chapter 6, the Barnwell Nuclear Fuels Plant (BNFP) Separations Facility is used to represent fuel reprocessing plants because detailed information is available. This facility is designed to recover uranium and plutonium from spent LWR fuel. Refer to Figure 6-1 for the plant flowsheet. Dissolution of the chopped fuel is followed by solvent extraction using tributyl phosphate (TBP) in a hydrocarbon diluent. The resultant product streams contain separated uranium and plutonium, and waste streams of high level, intermediate level and low level liquid wastes. Finally, there is considerable off-gas containing radioactive substances to be removed. Many are controlled by existing systems described in Chapter 6. This chapter attempts to predict levels of carbon-14 in various stages of the process so control mechanisms may be designed.

7.1 Carbon-14 in arriving spent fuel

Used fuel arrives and is placed in the Fuel Receiving and Storage Pool. The fuel arrives from many power stations, so the physical storage methods are quite flexible, and burnup of the fuel varies requiring adjustment of plant processes for each batch. It is assumed here that fuel has been utilized to an average burnup of 33,000 MWt-days per MTHM. The amount of carbon-14 generated in reactor fuel was computed according to methods and assumptions presented in Chapter 2. Results, given in units of curies per gigawatt (electric)-year, are listed in Table 2-1. The carbon-14 production rate from fuel irradiation is estimated to be 21.4 Ci/GWe-yr and it was assumed that the fuel turnaround is 33.5 MT per GWe-yr, so a rough estimation of incoming fuel-borne carbon-14 activity is 0.64 Ci/MT of fuel. No reduction of this value is assumed due to failed fuel losses, negative deviations in neutron fluxes in the reactor or the 150 day storage delay at the reactor sites prior to shipment to the reprocessor. No significant amount of the carbon-14 arriving at the Separations Facility is assumed to escape to the Fuel Storage Pool, even with some failed fuel. Therefore, based on 1,500 MT/year capacity at BNFP, approximately 960 curies per year of carbon-14 are assumed to reach the shearing-dissolving step, or head-end, of the plant.

7.2 Carbon-14 and stable carbon in the process pathways

The first step in nuclear fuel reprocessing is shearing. Fuel elements are mechanically transferred from the spent fuel pool to the remote process cell in which chopping occurs. They are chopped into segments 2 to 5 inches long, allowing hulls, oxide pellets and gases to be delivered to the dissolver. Hulls and pellets fall by gravity, while a sweep-air flow down the shear outlet chute passes off-gases to the dissolver.

In the dissolver, the oxide pellets are dissolved along with their impurities. Hulls are lifted from the dissolver, scrubbed and the wash solution and off-gas returned to the dissolver. It is assumed for this study that carbon, in whatever form it resides in the fuel, will be oxidized to carbon dioxide. Confirmation of this assumption awaits necessary laboratory data and/or actual field measurements at a reprocessing facility.

In Chapter 2, it was assumed that activation of oxygen-17 in the oxide fuel and of nitrogen-14 present as an impurity in the fuel (to the extent of 20 ppm) accounted for all carbon-14 formed therein. In this report, complete conversion to a gaseous state is assumed, releasing 0.64 Ci/MT of fuel processed to the dissolver off-gas system. At BNFP, then, 960 Ci/year needs to be removed from the off-gas system. Assuming a good dissolution and residence time of solution in the dissolver, nearly all the C-14 may be expected to be released to the dissolver off-gas system. The CO_2 concentration of the DOGS gas stream is assumed to be essentially that of air. The stream is not dry, however. In this design, the carbon dioxide concentration is taken to be 0.0315 percent by volume and the carbon-14 concentration is 3.6×10^{-6} Ci/scf based on a 300 day per year operating schedule. Other headend designs are possible which would produce a more concentrated carbon-14 off-gas stream, though details have not been finalized.

The assumptions used here are based on considerations lacking C-14 direct measurements. If carbon-14 were carried off in the liquid process streams, a small fraction of that in the dissolver might later volatilize in vessel overheads. It would be added to the off-gas stream called the composite VOGS and be entrained in a gas flow of nearly 3,200 scfm. Also, overheads.from the HLLW off-gas condenser might also contain a small amount of activity, which would be added to the composite VOGS to create a flowrate of 4,400 scfm. The concentration of carbon-14 in this stream would equal the total 960 Ci/year assumed to reside in the DOGS system; recall that the DOGS and VOGS combine to form a single stream for final iodine treatment and release. The CO_2 concentration is slightly higher than air in the final stream, however, because sugar is added in the high level liquid waste concentrator for denitration and ruthenium suppression, resulting in a considerable (non-radioactive) carbon dioxide contribution to the off-gas.

For this report, significant removal of carbon-14 is expected by treating the flow of the DOGS. It can be seen at BNFP that in the case of iodine, for example, treatment is applied once to the concentrated DOGS stream and once to the dilute but possibly contaminated VOGS stream. While absolute knowledge of carbon-14 contamination levels in the VOGS awaits measurement, it is felt that decontamination factors of 100 to 1,000 can be achieved by treating the DOGS stream. It is virtually assured that a last possible source of carbon-14 volatilization, the HLLW calciner, will provide a miniscule contribution to the total off-gas levels of this nuclide.

Chapter 7 References

1. Allied Gulf Nuclear SErvices. Barnwell Nuclear Fuel Plant Separation Facility Final Safety Analysis Report, Section 4, October, 1973.

2. Stephenson, M.J. and R.S. Eby. "Development of the FASTER Process for removing Krypton-85, carbon-14 and other contaminants from the off-gas of fuel reprocessing plants". Proceedings of the Fourteenth ERDA Air Cleaning Conference, August, 1976, (in publication).

CHAPTER 8. MODIFIED REPROCESSING PLANT EFFLUENT TREATMENT SYSTEMS

8.1 Cost estimates for separations facility off-gas treatment possibilities

In the following analysis, iodine removal is assumed to come first, followed by NO_x removal. Next comes C_{14} removal and Kr-85 removal. Iodine is removed first so as not to contaminate the $CaCO_3$ while NO_x removal is a necessary pretreatment for C_{14} and Krypton control

Due to the presence of other radionuclides, notably the high dose rate from Kr85, the process equipment is enclosed in process cells. At least two-foot thick concrete walls. additional Class I structure requirements, and radiation shielding constitute additional costs not included in the cost module that has been used throughout the report. Appropriate inclusions will be made.

Hot cell costs are sunk to the other radionuclide control systems which are much more expensive than the one for C_{14}. They are thus not included in the economic analysis.

As shown in Chapter 7, essentially all of the C-14 that is released from the fuel reprocessing operation passes through the dissolver off-gas system. By passing the dissolver off-gas stream through a caustic scrubbing system similar to those developed in previous chapters for LWR off-gas treatment, the desired removal of C-14 can be achieved. The dissolver off-gas stream at BNFP has a composition essentially the same as air. It has been estimated that an upper bound flow-rate, at operating temperature and pressure, of 740 cfm can be encountered in the Barnwell DOG stream. This flow-rate is considerably larger than those encountered in LWR systems;

therefore, a larger scrubbing system is needed. The system design is outlined in Appendix B.

From Appendix B, it can be seen that by cascading equal size absorption columns in series, desired removal of C-14 can be achieved. Due to the amount of $CaCO_3$ generated cartridge filters will not prove economical. Therefore, it is assumed that the system contains a backflush filter. As in Chapter 4, the following cases are defined:

Case I - 90% removal of CO_2 (1 column)
Case II - 99% removal of CO_2 (2 columns)

In case II, where more than 1 column is used, additional pumps are needed. Also, because of the design flowrates of the liquid and gas entering the scrubber, the system will contain 2 inch, 304 stainless steel piping throughout.

Capital costs for a 740 cfm dissolver off-gas removal system for C-14

Table 8-1 summarizes the equipment costs for the dissolver off-gas C-14 removal system. Cost data for this larger system were obtained from the sources referenced in Chapter 4.

As indicated in Chapter 4, the fabricated equipment costs are assumed to represent 13.3 percent of the capital cost of the installed system. Therefore, capital costs of each case are as follows:

Case I	$600,000
Case II	728,000

These estimates do not include:
a) - shielding and other Class I structure requirements
b) - loss of production during installation

c) - major retro-fit costs which involve changes
to space envelopes

d) - licensing activities and delays.

Item a will be estimated and added to the above capital costs. These cost items will be in approximately the same cost range as indicated for reactors in Chapter 4. The cost of NO_x removal is assumed to be borne by the Krypton removal system because the latter is required by EPA and also needs NO_x pretreatment. Therefore no costs for NO_x removal are included in the economic evaluation of C_{14} control in reprocessing plants.

For CO_2 removal by caustic scrubbing, a generic process cell of dimensions 12' x 12' x 20' is considered for costing purposes. Due to the presence of Krypton-85 and other radionuclides, two-foot thick concrete walls, steel linings and reinforced Class I structures are required.

The additional unit volume cost, after escalation to 1977, is $\$114/ft^3$ (1). The capital costs of the process cell facilities is $328,000.

It is felt that the retrofit costs should be charged off to the control of Krypton and other radionuclides, because the C_{14} subsystem is only a minor process component in the integrated radionuclide control case. Therefore, the total capital costs for the two cases are:

Case I - $600,000 + $328,000 = $ 928,000
Case II - $728,000 + $328,000 = $1,056,000

Annual CO_2 fixation

The annual operating costs for the 740 cfm system include labor, chemicals, utilities, maintenance, and replacement. Maintenance and replacement costs included repair and replacement of equipment. For this study, 7 percent of the

Table 8-1

Fabricated Equipment Costs for Reprocessing Dissolver Off-Gas Scrubbing System (740 cfm)

Item		Cost
1. Off-Gas Blower		$ 2,800
2. Absorption Column(s)	- Case I	12,000
	- Case II	24,000
3. Column Internals		
a. Packing	- Case I	1,700
	- Case II	3,400
b. Gas Injection Support Plates		
	- Case I (2)	800
	- Case II (4)	1,600
c. Liquid Distributor		
	- Case I	400
	- Case II	800
d. Wire Mesh		50
e. Wash Down Nozzle		70
4. Mixing Tank		720
5. Agitator		750
6. Chemical Charging Tanks		
a. NaOH		350
b. $Ca(OH)_2$		350
7. Recirculation Pump	- Case I	1,200
	- Case II	2,400
8. Filter Pump		1,200

Table 8-1 (Continued)

Item	Cost
9. $CaCO_3$ Backflush Filter Unit	$58,000
Total fabricated equipment cost	
a. Case I	$80,000
b. Case II	97,000

original capital cost is assumed for these costs. An average capital cost of $992,000 is assumed for the preceding two cases. The labor cost assumes one additional staff member at the qualification level of an operator at $25,000 per man-year. Chemical costs are assumed to be $2,000 per year with negligible utility costs.

Labor	
$25,000 x 1.0 =	$ 25,000
Maintenance and Replacement	
$992,000 x 0.07	69,000
Chemicals	2,000
Fixed Charges	198,000
Annual Costs	$294,000

The 20-year present worth factor at 8 percent interest per year is 9.82. Therefore, the present worth value of the 20 years of annual costs would be 9.82 x $294,000 $\cong$ $2,887,000.

Since Barnwell is an older design the radiation protection standards and guides were much less restrictive at its inception than those currently required. It is evident that in future reprocessing plants the DOG flow rates will be drastically reduced. For this reason a lower flow of 100 cfm for the DOG is also considered, to reflect future designs and provide an associated range of costs.

Capital costs for a 100 cfm dissolver off-gas removal System for C-14

Appendix C reflects the design for a caustic scrubbing unit to treat a 100 cfm DOG flow rate. Once again the following Cases are defined:

Case I - 90% removal of CO_2
Case II - 99% removal of CO_2

The system will contain 2 inch, 304 stainless steel piping throughout, and due to waste volumes generated, only the backflushable option will be considered. Table 8-2 summarizes the fabricated equipment costs for this system, as obtained from the sources referenced in Chapter 4. Using the same procedure as that for the 740 cfm system with a process cell cost equal to 2/3 of the 740 cfm case, the total capital costs for each case are as follows:

Case I - $399,000
Case II - 421,000

Annual fixation costs

Using an average installed cost of $410,000 for the above two cases the annual costs are:

Labor	
$25,000 x 1.0 =	$25,000
Maintenance and Replacement	
$410,000 x .07	$29,000
Chemicals	$1,500
Fixed charges	$82,000
Annual Costs	$137,000

The 20 year present worth factor is

9.82 x 137,000 = $1,345,000

Table 8-2

Direct Equipment Costs for Reprocessing Dissolver Off-Gas Scrubbing System (100 cfm)

Item		Cost
1. Off-Gas Blower		$850
2. Absorption Column(s)	- Case I	$3,900
	- Case II	$6,300
3. Column Internals		
a. Packing	- Case I	$240
	- Case II	$480
b. Gas Injection Support Plates		
	- Case I (1)	$280
	- Case II (2)	$560
c. Liquid Distributor		
	- Case I	$290
	- Case II	$290
d. Wire Mesh		$30
e. Wash Down Nozzle		$50
4. Mixing Tank		$500
5. Agitator		$650
6. Chemical Charging Tanks		
a. NaOH		$350
b. $Ca(OH)_2$		$350
7. Recirculation Pump		$1,200
8. Filter Pump		$1,200

Table 8-2 (Continued)

Item	Cost
9. $CaCO_3$ Backflush Filter Unit	$17,000
Total fabricated equipment cost	
a. Case I	$24,000
b. Case II	$27,000

8.2 Effect of integrated control technologies

Other off-gas control systems may be applied at nuclear fuel reprocessing plants which will have a bearing on Carbon-14 control. Specifically, control of noble gas fission products has been proposed using a fluorocarbon absorbent or a cryogenic distillation device. It was shown in Chapter 3 that these systems presently require pre-treatment to remove fouling agents such as carbon dioxide. The devices to accomplish this task are an integral part of the noble gas control system.

No reliable evidence is available on which to base an assessment of the cold-trap as a means of CO_2 transfer to another device which yields a solid product form.
It is felt that such a system could be created given the demand, though none has been designed to date. On the other hand, it has been shown that another likely interceptor, molecular sieves, may be applicable as a CO_2 pre-treatment system. Speculation regarding impacts to the combined cost for carbon-14 and noble gas isolation are therefore summarized:

- The total cost of carbon-14 treatment is somewhat insensitive to scrubber size, and greatly dependent on the mass of CO_2 treated, so eventual costs will not deviate significantly from estimates for this reason alone.
- Due to the immature nature of these technologies, with respect to demonstration at commercial facilities, conservation is appropriate and suggests that computed credits due to system cost coupling may have limited demonstrable significance. Conceptually, savings can be achieved by the use of common hot cell space and associated equipment as well as sunk costs due to the sharing a NO_x removal system.

Chapter 8 References

1. J. T. Long, Engineering For Nuclear Fuel Reprocessing, Gordon and Breach Science Publishers, New York, 1967. p. 933.

CHAPTER 9. WASTE MANAGEMENT OPTIONS FOR CARBON-14 PRODUCT FORMS

9.1 Introduction

The preceding chapters have addressed technologies and costs associated with the control and capture of carbon-14. The next logical question is the disposition of the final carbon-14 waste product form.

The criteria and regulations relative to waste management of light water reactor fuel cycle wastes have yet to be formulated. Nonetheless, some projections as to the nature of the operations and costs can be made based on present waste management trends.

9.2 Disposal

There appear to be two major options for the disposal of captured C-14. These options are shallow-land burial and deep geological emplacement. There is a marked difference in cost between these two methods. Future regulations based on health, safety, and environmental evaluations will dictate which disposal method must be utilized.

9.3 C-14 product form and package

Carbon-14 is captured as calcium carbonate by filtration. Two filtration options have been examined. The first option is to collect the calcium carbonate cake on disposable filter cartridges. The second option is to collect it on an etched disc filter which can be backflushed.

In the first option, the loaded filter cartridges are handled as solid waste and are disposed of by being concreted in 55-gallon drums. In the second option, the disc filters are backflushed and the resultant slurry is incorporated into concrete by a radwaste solidification unit. The concrete matrix for the calcium carbonate is then packaged in 55-gallon drums.

No acute radiological hazard will result from exposure to carbon-14 in the packaged product, though it is assumed handling will be done remotely prior to concrete fixation. Estimates of stable carbon to carbon-14 ratios (expressed as ratios of stable carbon atoms (N_S) to radioactive carbon atoms (N_R) in the product material) are summarized below:

- BWR (9.0 Ci/year, 7968 g-mole/year stable carbon)

  $$\frac{N_S}{N_R} = 5.5 \times 10^4$$

  mole fraction $^{14}C = 1.8 \times 10^{-5}$

- PWR (3.0 Ci/year, 398 g-mole/year stable carbon)

  $$\frac{N_S}{N_R} = 8.2 \times 10^3$$

  mole fraction $^{14}C = 1.2 \times 10^{-4}$

- Separations Facility - 740 cfm process rate (960 Ci/year, 1.48×10^5 g-mole/year stable carbon)

  $$\frac{N_S}{N_R} = 9.6 \times 10^3$$

  mole fraction $^{14}C = 1.0 \times 10^{-4}$

- Separations Facility - 100 cfm process rate (960 Ci/year, 1.99×10^4 g-mole/year stable carbon)

  $$\frac{N_S}{N_R} = 1.29 \times 10^3$$

  mole fraction $^{14}C = 8.0 \times 10^{-4}$

9.4 BWR waste management

9.4.1 Waste volumes and cost calculations for BWR's

Option A - Disposable cartridge filters

Case 1. Surface dose <200mR/hr.

Assumes:

- $0.40/gal concretion cost
- eighteen cartridge filters per drum
- 300 operating days per year
- 1,016 drums of concreted filters per year
- a final radwaste product density of 100 lbs/ft^3
- 1600 lbs shielding/42000 lbs payload per truck in shipment to disposal site(1)
- $2.13 per 100 lbs load
- 500 miles to disposal site
- disposal costs of $1.30 per cubic foot for shallow-land burial(1)
- disposal costs of $24.50 per rectilinear cubic foot for deep geological emplacement(1)
- new 55-gal drum are $15 each(1)
- concretion waste containers (drums), transportation, and disposal are the significant costs
- no other radionuclide present

The calculations are as follows:

concretion cost: $0.40/gal x 1016 drums/yr x 55 gal/drum = $24,000 per year

cost of drums -

1,016 drums/yr x $15/drum = $15,200 per year

cost of transportation -

(1600 lbs shielding + 1,016 drums/yr x 750 lbs/drum) x $2.13/100 lbs = $16,300 per year

cost of disposal (shallow land burial) -
1,016 drums/yr x 7.5 ft^3/drum x \$1.30/ft^3 = \$9,900 per year

total annual waste management charges -
concretion - \$22,400
drums - 15,200
transportation - 16,300
disposal - 9,900
\$64,000 per year

Assuming a 30-year life at 8 percent interest, the present worth value equals

11.26 x \$64,000 = \$720,000

If it is determined that shallow land burial is not an acceptable means of disposal for long-lived isotopes such as C-14, the cost of disposal becomes:

cost of disposal (deep geological emplacement) -
1,016 drums/yr x 12 ft^3/drum* x \$24.50 = \$299,000

As can be seen in the case of deep geological emplacement, the disposal costs greatly exceed all other operational costs and the total waste management charges become:

concretion - \$ 22,400
drums - 15,200
transportation - 16,300
disposal - 299,000
\$353,000 per year

Again, assuming a 30-year life at 8 percent interest, the present worth value equals

11.26 x \$353,000 = \$3,975,000

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* Note: The volume of a 55-gal drum is 7.5 ft^3 but the rectilinear space required to store it is 12 ft^3.

Case 2. Surface dose rate >200 mR/hr

- concretion cost of \$0.60/gal
- disposal costs of \$3.20/ft^3 for shallow land burial
- shielding weight of 39,500 lb/42,000 lb of waste
- other assumptions as in Case 1

cost of concretion: \$0.60/gal x 1,016 drums/year x 55 gal/drum = \$33,500

cost of transportation = cost of transportation of wastes
+ cost of transportation of shielding

Waste weight

1016 drums/yr x 750 lbs/drum = 762,000 lbs/yr

Shielding weight -

$$762{,}000 \text{ lbs waste} \left(\frac{39{,}500 \text{ lbs shielding}}{42{,}000 \text{ lbs waste}}\right) = 717{,}000 \text{ lbs}$$

Transportation Cost:
(762,000 + 717,000)lbs. x \$2.13/100lbs = \$31,500 per year

Cost of disposal:

1016 drums/yr x 7.5ft^3/drum x \$3.20/ft^3 = \$24,400/year

Total annual waste management charges

concretion	\$33,500
drums	15,200
transportation	31,500
disposal	24,400
	\$105,000 per year

Present worth value

11.26 x \$105,000 = \$1,182,000

Cost of disposal (deep geological emplacement): \$299,000
(as in Case 1)

concretion	\$ 33,500
drums	15,200
transportation	31,500
disposal	299,000
	\$379,000

Present worth value:

11.26 x \$379,000 = \$4,268,000

Option B - Backflushable etched disc filters

Case 1. Surface dose rate <200mR/hr

Assumes:

- Solidification at \$0.40/gal (increase in utilities and labor at radwaste solidifier)
- 300 operating days per year
- a final radwaste product density of 100 lbs/ft^3
- 42,000 lbs payload per truck shipment to disposal site
- \$2.13 per 100 lbs load
- 500 miles to disposal site
- disposal costs of \$1.30 per cubic foot for shallow-land burial
- disposal costs of \$24.50 per rectilinear cubic foot for deep geological emplacement
- new 55-gal drums are \$15 each
- waste containers (drums), transportation, and disposal are the significant costs
- at a 99 percent recovery of the incoming CO_2, 1,410 lbs of $CaCO_3$ are filtered per year
- the filters must be backflushed with 3 gallons of water each time 2.2 lbs of filtrate is collected on the filter
- the backflush slurry is incorporated into concrete at a ratio of 30 percent by weight to obtain a final radwaste product
- 1600 lb of shielding required for 42,000 lb waste

The calculations are as follows:

pounds per year of radwaste slurry -

$$\left(\frac{1{,}410}{2.2}\right)(3)\ (8.3) \text{ lbs of flush solution} + 1{,}410 \text{ lbs of } CaCO_3 = 17{,}400 \text{ lbs per year}$$

pounds per year of final concrete product -

$$\frac{17{,}400}{.3} = 58{,}000 \text{ lbs per year}$$

shielding weight required: $\left(\frac{1600}{42000}\right)$ (58,000) = 2200 lbs per year

number of 55-gallon drums per year -

$$\frac{58{,}000 \text{ lbs/year}}{(100 \text{ lbs/ft}^3)(7.5 \text{ ft}^3\text{/drum})} = 77 \text{ drums}$$

concretion cost: $0.40/gal x 77 drums/yr x 55 gal/drum =
$1700 per year

cost of drums -

77 x $15 = $1,200 per year

cost of transportation -

$2.13/100 lb x (58,000 lbs + 2,200 lbs) = $1300/year

cost of disposal (shallow-land burial) -

77 x 7.5 x $1.30 = $750 per year

total waste management charges -

solidification and concretion	-	$1,700
drums	-	1,200
transportation	-	1,300
disposal	-	750
		$5,000 per year

Assuming a 30-year life at 8 percent interest, the present worth value equals

11.26 x $5,000 = $56,000

If deep geological emplacement is used to dispose of the waste, the disposal costs become

77 drums/year x 12 ft^3/drums x $24.50/$\text{ft}^3$ = $22,600 per year

The total waste management charges are:

solidification and concretion	-	$ 1,700
drums	-	1,200
transportation	-	1,300
disposal	-	22,600
		$27,000 per year

Again, assuming a 30-year life at 8 percent interest, the present worth value equals

$27,000 x 11.26 = $304,000

Case 2: Surface dose rate >200mR/hr

Assume:

- \$0.60/gal for solidification and concretion
- disposal costs of \$3.25/ft^3 for shallow land burial
- 39,500 lb of shielding/42000 lbs of waste

. solidification and concretion \$0.60/gal X 77 drums/year x 55 gal/drum = \$2,500

Total transportation costs:

$$58000 \text{ lbs} + \left(\frac{39,500}{42,000}\right) 58,000 \text{ lbs} \quad \frac{\$2.13}{100 \text{ lbs}} = \$2,400$$

cost of disposal:

\$3.25/ft^3 x 7.5 ft^3/drum x 77 drums/year = \$1,900/year

Total waste management charges:

solidification and concretion	\$2,500
drums	1,200
transportation	2,400
disposal	1,900
	\$8,000

Present worth value:

11.20 x \$8000 = \$90,000

Deep geological emplacement costs: \$22,600 per year

Total waste management charges: \$29,000

Present worth value:

\$29,000 x 11.26 = \$327,000

9.4.2 Waste management cost summary for BWR systems

There are two options for filtration and two disposal cases for each option. The following tabulated values are the 30-year present worth values.

Option A (Disposable Cartridge Filters)

	Case 1 (low dose rate)	Case 2 (high dose rate)
Shallow Land Burial	\$ 720,000	\$1,182,000
Deep Geological Emplacement	\$3,975,000	\$4,268,000

Option B (Backflushable Filters)

	Case 1 (low dose rate)	Case 2 (high dose rate)
Shallow Land Burial	$ 56,000	$ 90,000
Deep Geological Emplacement	$304,000	$327,000

As can be seen, the cost of waste management can range over two orders of magnitude depending on which option is chosen.

9.5 PWR Waste management

The total volume of off-gas treated by a PWR carbon-14 collection system is approximately 5 percent of that treated by a BWR carbon-14 system. This directly affects the volume of calcium carbonate generated and the amount of waste requiring both transport and disposal. Calculations for a PWR are based on the same methods and assumptions used in the BWR case. The number of filters used and amount of backwashed solution is 5 percent of that found in a BWR. The waste management cost summary for DWR systems is:

	Case 1 (low dose rate)	Case 2 (high dose rate)
Option A (Disposable Cartridge Filters)		
Shallow Land Burial	$ 36,000	$ 59,000
Deep Geological Emplacement	$199,000	$213,000
Option B (Backflushable Filters)		
Shallow Land Burial	3,000	4,500
Deep Geological Emplacement	$15,000	$16,000

9.6 Spent LWR fuel reprocessing plant waste management

The design and design parameters for a carbon-14 off-gas cleanup system for nuclear fuel reprocessing facilities are discussed in Chapter 8. Because of the relatively large volume of calcium carbonate generated by the reference system only the backflushed filter system is viewed as an option. The assumptions for the volume and cost calculations are essentially the same as listed for the BWR carbon-14 cleanup system.

For a 740 cfm dissolver off-gas cleanup system 25,900 pounds of calcium carbonate are generated per year. This requires approximately 35,000 gallons of backflush solution. The resultant calcium carbonate slurry weighs approximately 319,000 pounds.

Assuming a matrix for the calcium carbonate slurry of 30 percent slurry and 70 percent concrete a final radwaste product of 1,060,000 pounds per year is generated. At 750 pounds per drum of radwaste, 1,410 drums of concreted calcium carbonate would be generated each year.

Case 1: Dose rate <200mR/hr

The cost calculations are as follows:

Solidification and concretion costs -

$0.40/gal x 1410 drums/yr x 55 gal/drum = $31,000 per year

drum costs -

1,410 x $15 = $21,200 per year

shielding required -

$$1{,}060{,}000 \text{ lbs} \times \frac{1600 \text{ lbs shield}}{42000 \text{ lbs waste}} = 40{,}400 \text{ lbs per year}$$

transportation costs -

$$(40{,}400 + 1{,}060{,}000)\frac{\text{lbs}}{\text{year}} \times \frac{\$2.13}{100\text{lbs}} = \$23{,}500 \text{ per year}$$

cost of disposal (shallow-land burial) -

$1{,}410 \text{ drums/year} \times 7.5 \text{ ft}^3\text{/drums} \times \$1.30/\text{ft}^3 = \$13{,}700$ per year

total waste management charges -

solidification and concretion	-	$31,000
drums	-	21,200
transportation	-	23,500
disposal	-	13,700
		$89,000 per year

Assuming a 20-year operating life for a reprocessing facility at 8 percent interest, the present worth of these operating costs would be

9.82 x $89,000 = $874,000

If deep geological emplacement is required for the disposal of this waste form, the disposal costs become:

$1{,}410 \text{ drums/year} \times 12 \text{ ft}^3\text{/drums} \times \$24.50/\text{ft}^3 = \$415{,}000$

The total waste costs become:

solidification and concretion -	$ 31,000
drums	21,200
transportation	23,500
disposal	415,000
	$491,000 per year

Again, assuming a 20-year life at 8 percent interest, the present worth value equals

9.82 x $491,000 = $4,822,000

Case 2: Dose rate >200mR/hr

The cost calculations are as follows:

solidification and concretion costs -

$0.60/gal x 1410 drums/yr x 55 gal/drum = $46,500 per year

shielding required -

$$1{,}060{,}000 \text{ lbs} \times \frac{39{,}500}{42{,}000} = 997{,}000 \text{ lbs per year}$$

transportation cost -

$$(997{,}000 \times 1{,}060{,}000) \text{ lbs} \times \frac{\$2.13}{\text{lbs}} = \$43{,}800 \text{ per year}$$

cost of disposal -

$$1{,}410 \text{ drums/year} \times 7.5 \text{ ft}^3\text{/drum} \times \$3.25\text{/ft}^3 = \$34{,}400 \text{ per year}$$

Total waste management charges:

solidification and concretion	-	$46,500
drums	-	21,200
transportation	-	43,800
disposal	-	34,400
		$146,000 per year

Present worth value:

9.82 x $146,000 = $1,434,000

Deep geological emplacement costs: $415,000 as in Case 1

Total waste management costs: $527,000 per year
x 9.82 = Present worth value: $5,175,000

In summary, the present worth values of the above cases are:

	Case 1 (<200mR/hr)	Case 2 (>200mR/hr)
Shallow land burial	$ 874,000	$1,434,000
Deep geological Emplacement	$4,822,000	$5,175,000

For a 100 cfm dissolver off-gas cleanup system, all costs are a direct ratio of 100 cfm to 740 cfm. These are:

	Case 1 Surface dose rate <200mR/hr	Case 2 Surface dose rate >200mR/hr
Shallow land burial		
Total waste management charges	- $ 12,000/year	$ 20,000/year
Present worth	- $118,000	$194,000
Deep geological emplacement		
Total waste management charges	$ 66,000/year	71,000/year
Present worth	$650,000	$697,000

1. Croff, A.G., An Evaluation of Options Relative to the Fixation and Disposal of C-14 Contaminated CO_2 and $CaCO_3$, April 1976, ORNL TM-5171.

CHAPTER 10. ECONOMIC COMPARISONS AND SUMMARY

In the preceding chapters of this report, the sources of carbon-14 in LWR's and fuel reprocessing facilities have been identified. Also, systems for the removal of carbon-14 in existing plants and future plants have been addressed from both a technological and an economic standpoint. Within existing technology, it is shown that caustic scrubbing is the most cost-effective alternative for concurrently removing C-14 from waste-gas streams and leaving it in a form compatible with permanent disposal conditions

Table 10-1 gives a summary and comparison of costs for a carbon-14 removal system for BWR's, PWR's and fuel reprocessing facilities. In BWR's the total installation cost for a scrubbing system with backflushable filters is greater than one with disposable cartridge filters. However, upon examination of the waste management costs, it can be seen that the back-flushable filter scrubbing system more than pays for itself over a thirty-year plant lifetime, since these costs are a factor of ten lower than using cartridge filters. The same can be said for PWR waste management costs.

Because of the possibility that a recombiner will have to be installed in a PWR off-gas system, the total capital costs for a PWR scrubbing system for C-14 removal are on the order of a factor of four greater than a BWR system and approaching that of an existing reprocessing facility system.

It can be seen from Table 10-1 that should both shallow land burial and deep geological emplacement be deemed acceptable methods of permanent disposal, shallow land burial is a factor of ten less expensive than deep geological emplacement. The relative differences between high and low activity waste disposal costs become small for deep geological placement when compared with the magnitude of the costs involved.

In conclusion, it should be reemphasized that the preceding cost evaluation did not include items like loss of production during installation, retro-fitting costs when modifications must be made to the physical plant, and costs incurred from licensing actions. These factors are deemed to be so variable, and so often defy prediction, that a fair comparison of costs could not be made. Nonetheless, modifications and licensing will require time, labor and materials, and surely cause some loss of production. The range of costs is presented:

Cost Factors Directly Related to Retrofit

	Range $ x 10^6
Lost Production Revenue ranging from 1 week to 1 month	2 to 15
Licensing activity costs	0.1 to 1.0
Unusual construction problems	0.05 to 0.5

	BWR		PWR (Note 1)		Reprocessing Plant	
	Cartridge Filter	Backflush Filter	Cartridge Filter	Backflush Filter	Backflush Filter	
					740cfm	100cfm
Total Capital Costs						
90% removal of C-14	$ 67,500	$ 128,000	$ 855,000	$ 855,000	$ 928,000	399,000
99% removal of C-14	$ 75,000	$ 143,000	$ 855,000	$ 855,000	$ 1,056,000	421,000
Annual CO_2 Fixed Costs	$ 53,000	$ 53,000	$ 257,000	$ 257,000	$ 294,000	137,000
Present Worth of Annual Fixed Costs	$ 597,000	$ 597,000	$ 2,894,000	$2,894,000	$ 2,887,000	1,345,000
Annual Waste Management costs						
- shallow land burial						
low activity	$ 64,000	$ 5,000	$ 3,200	$ 300	$ 89,000	12,000
high activity	$ 105,000	$ 8,000	$ 5,200	$ 400	$ 146,000	20,000
- deep geological emplacement						
low activity	$ 353,000	$ 27,000	$ 18,000	$ 1,400	$ 491,000	66,000
high activity	$ 379,000	$ 29,000	$ 19,000	$ 1,500	$ 527,000	71,000
Present Worth of Waste Management Costs						
- shallow land burial						
low activity	$ 720,000 (30 yr)	$ 56,000 (30 yr)	$ 36,000 (30 yr)	$ 3,000 (30 yr)	$ 874,000 (20 yr)	118,000 (20 yr)
high activity	$1,182,000 (30 yr)	$ 90,000 (30 yr)	$ 59,000 (30 yr)	$ 4,500 (30 yr)	$ 1,434,000 (20 yr)	194,000 (20 yr)
- deep geological emplacement						
low activity	$3,975,000 (30 yr)	$ 304,000 (30 yr)	$ 199,000 (30 yr)	$ 15,000 (30 yr)	$ 4,822,000 (20 yr)	650,000 (20 yr)
high activity	$4,268,000 (30 yr)	$ 327,000 (30 yr)	$ 213,000 (30 yr)	$ 16,000 (30 yr)	$ 5,175,000 (20 yr)	697,000 (20 yr)

TABLE 10.1 SUMMARY AND COMPARISON OF COSTS

NOTE 1. The C-14 recovery efficiency expressed here is limited to C-14 recovery from the primary rad-waste gas system.

APPENDICES

Nomenclature

A	Cross sectional area of column, ft^2.
a	Specific Packing Surface, ft^2/ft^3.
G	Gas rate, lb/ft^2-sec.
G'	Gas mass flow rate, lb/hr.
G_F	Gas flooding velocity, lb/ft^2-sec.
g_c	Gravatational Constant, 32.2 ft/sec^2.
h	Height of column, ft.
$K_G a$	Mass transfer coefficient, lb-mole/hr-ft^3-atm.
L	Liquid rate, lb/ft^2-sec.
L'	Liquid Mass flow rate, lb/hr.
N	lb-moles of desired gas transferred per hour.
ΔP_{LM}	Log mean partial pressure drive, dimensionless
ϵ	Porosity (bed voidage), dimensionless
μ	Viscosity of liquid, centipoise.
ρ_G	Density of Gas, lb/ft^3.
ρ_L	Density of liquid, lb/ft^3.
ψ	Ratio of water density to that of liquid in column, dimensionless

APPENDIX A - LWR SCRUBBER SYSTEM DESIGN

It is desired to remove CO_2 from an LWR off-gas system. The off-gas essentially has the composition of air and contains .0315% CO_2 by volume. It is flowing at 40 cfm. A packed column is to be designed for the absorption process, operating at atmospheric pressure and 20°C.

The following definitions, assumptions, and parameters will be used in the column design:

1. 1-inch Berl saddles will be used as packing.

2. The packing factor $F(p) = a/\epsilon^3 = 110$ for 1-inch Berl saddles (see reference 2, page 56), where "a" is the specific packing surface (ft^2/ft^3), and "ϵ" is the porosity.

3. A 2 N(8wt%) aqueous sodium hydroxide solution will be used. From reference 1, p. 3-78, the specific gravity of a 2 N NaOH solution at 20°C is 1.0869. The density, ρ_L, of the solution is:

$$\rho_L = 1.0869 \times 62.3 \frac{lb}{ft^3} = 67.7 \frac{lb}{ft^3}.$$

4. The average molecular weight of dry air is 28.97 $\frac{lb}{lb\text{-}mole}$. At 20°C (68°F), the density, ρ_G, of the off-gas will be

$$\rho_G = \frac{28.97 \frac{lb}{lb\text{-}mole}}{359 \frac{ft^3}{lb\text{-}mole}} \times \frac{(460 + 32)^\circ R}{(460 + 68)^\circ R} = .0752 \frac{lb}{ft^3}.$$

5. Let ψ = ratio of the density of water to that of the liquid in the column, so,

$$\psi = \frac{1}{1.0869} = .92.$$

6. From reference 4, p. C-26, the viscosity of an 8% NaOH solution at 20°C is 1.6 centipoise.

System Design

The off-gas flow rate is 40 ft^3/min. So, the mass flow rate, G′, is:

$$G' = 40\ \frac{ft^3}{min} \times .0752\ \frac{lb}{ft^3} \times \frac{60\ min}{hour} = 180.48\ \frac{lb}{hr}.$$

Try a liquid mass flow rate, L′, that is 81 times the gas flow rate. So,

$$81 \times 180.48 = 14{,}618.9\ \frac{lb}{hr} = 14{,}618.9\ \frac{lb}{hr} \times \frac{1\ ft^3}{67.7\ lb} \times 7.48\ \frac{gal}{ft^3} \times$$

$$\times \frac{1\ hr}{60\ min} = 26.9\ gpm.$$

Figure A-1 shows the generalized pressure drop correlation for packed tower design. The abscissa value is

$$\frac{L'}{G'}\left(\frac{\rho_G}{\rho_L}\right)^{\frac{1}{2}} = 81\left(\frac{.0752}{67.7}\right)^{\frac{1}{2}} = 2.7.$$

Now referring to Figure A-1, for a pressure drop of .25 inches of water/ft. of packing which is sound column design, the ordinate value is

$$F(G^2) = \frac{G^2\, a\, \psi\, \mu^{0.2}}{\rho_G \rho_L \epsilon^3\, g_c} \cong .00277 = \frac{G^2 \times 110 \times (.92)(1.6)^{.2}}{(.0752)(67.7)(32.2)} = G^2 \times 0.68.$$

so,

$$G = \left(\frac{.00277}{0.68}\right)^{\frac{1}{2}} = .0638\ \frac{lb}{ft^2\text{-}sec} = 229.68\ \frac{lb}{ft^2\text{-}hr}.$$

The diameter of the column will be:

$$\left(\frac{180.48}{229.68} \times \frac{4}{\pi}\right)^{\frac{1}{2}} = 1.0\ ft.$$

From Reference 3, p. 74, $K_G a \cong 2.25$ lb-mole/hr-ft^3-atm.

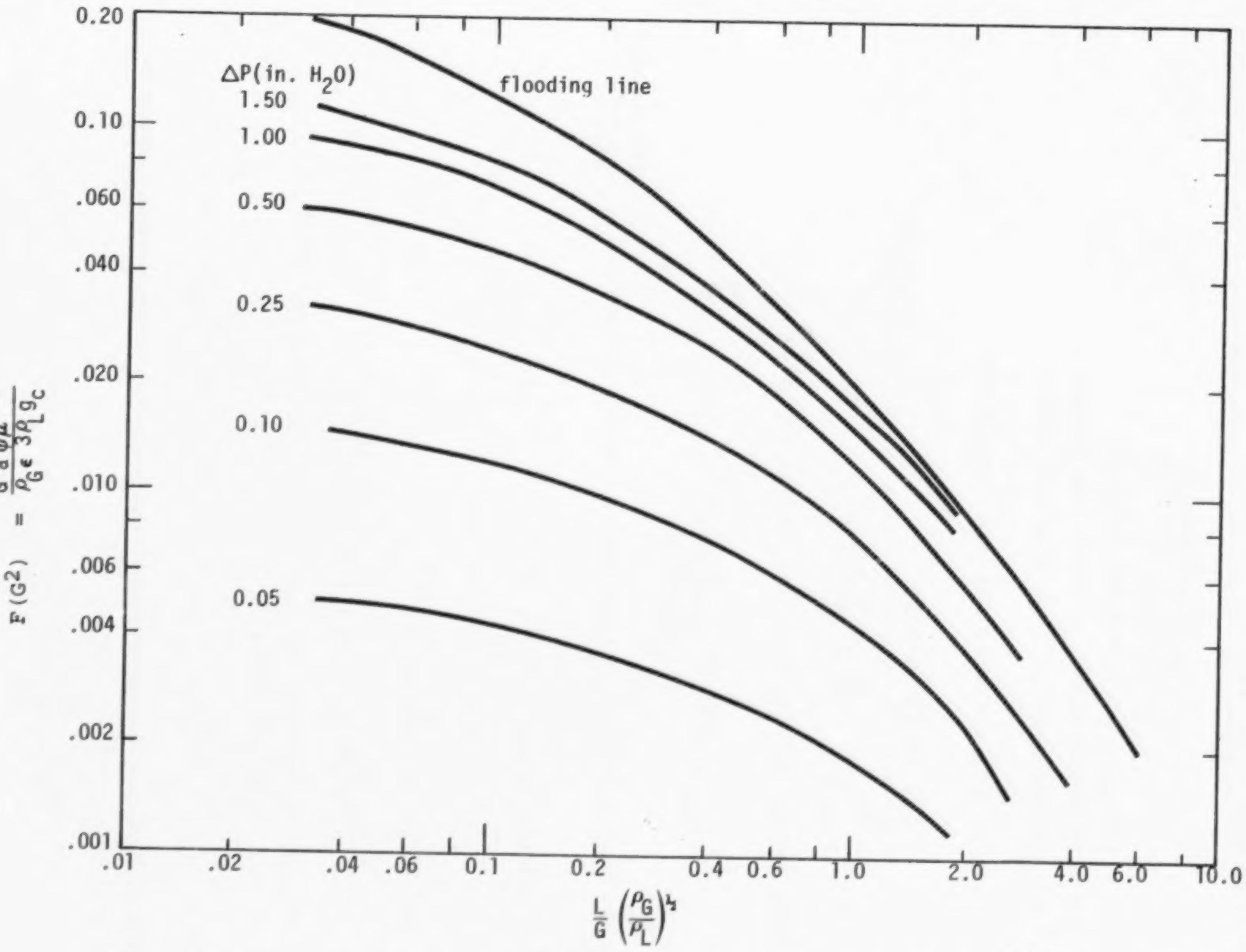

Figure A-1. Generalized pressure drop correlation for sizing packed towers.[2]

From Reference 2, p. 55,

$$K_G a = \frac{N}{h \quad A \quad \Delta P_{LM}}$$

where N = lb-moles CO_2 transferred/hr

A = cross sectional area of the column

ΔP_{LM} = log mean partial pressure drive.

The amount of CO_2 entering the absorber/hr is

$$\frac{180.48 \frac{lb}{hr} (.000315)}{\left(.0752 \frac{lb}{ft^3}\right)\left(359 \frac{ft^3}{mole}\right)\left(\frac{460 + 68}{460 + 32}\right)} = .00196 \frac{\text{lb-moles}}{\text{hour}}.$$

For 90% removal of CO_2 at 1 atm operating pressure

$$\Delta P_{LM} = \frac{.000315 - .0000315}{\ln\left(\frac{.000315}{.0000315}\right)} = .000123.$$

For 90% removal $\Rightarrow N = .9(.00196) = .001766$ lb-moles $\frac{CO_2 \text{ transferred}}{\text{hour}}$.

The cross-sectional area of the column is

$$A = \frac{\pi}{4} (1)^2 = .7854 \text{ ft}^2.$$

The height of the column is

$$h = \frac{.001766}{(2.25)(.7854)(.000123)} = 8.12 \text{ ft}.$$

The residence time of the liquid in the column packed with 1 inch Berl saddles ($\epsilon = .78$) is

$$(.7854 \text{ ft}^2 \times 8.12 \text{ ft} \times .78)/\left(26.9 \frac{gal}{min} \times \frac{1 \text{ ft}^3}{7.48 \text{ gal}}\right) = 1.38 \text{ minutes}.$$

Next the result is checked for flooding velocity. From Figure A-1, at flooding $F(G^2) = .0062$. So,

$$.0062 = G_F^2 \times .68 \Rightarrow G_F = .095 \frac{lb}{ft^2\text{-sec}}.$$

Now,

$$\frac{.0638}{.095} \times 100 = 67.2$$

So, operation is at 67.2% of flooding, which is sound column design.

Now, if 99% removal of CO_2 is desired

$$\Delta P_{LM} = \frac{.000315 - .00000315}{\ln\left(\frac{.000315}{.00000315}\right)} = .0000677$$

$$N = .99(.00196) = .00194$$

$$h = \frac{.00194}{2.25(.7854)(.0000677)} = 16.22 \text{ ft.}$$

The residence time of the liquid in the column is about twice that for 90% removal since the column is about twice as tall.

The column is operating at 67.2% of flooding.

Table A-1

Desired CO^2 Removal	Liquid Flow Rate	Gas Flow Rate	Column Diameter	Column Height	Residence Time	Operational % of Flooding
90%	26.9 gpm	40 ft^3/min	1 ft (12 inches)	8.12 ft	1.38 min	67.2%
99%	26.9 gpm	40 ft^3/min	1 ft (12 inches)	16.22 ft	1.38 min	67.2%

References

(1) Perry, R. H., Chilton, C. H., ed., Chemical Engineer's Handbook, Fifth Edition, McGraw-Hill Book Co., N. Y., 1973.

(2) Eckert, J. S., "Design Techniques for Sizing Packed Towers," Chemical Engineering Progress, Vol. 57, No. 9, September 1961.

(3) Eckert, J. S., "How Tower Packings Behave," Chemical Engineering, April 14, 1975.

(4) Fleming, R., A Compilation of Physical and Chemical Properties of Materials and Streams Encountered in the Chemical Process Department, Manual HW-57386, General Electric Company-Hanford Atomic Products Operation, 1958.

APPENDIX B - REPROCESSING DISSOLVER OFF-GAS SYSTEM DESIGN (CATEGORY I)

It is desired to remove CO_2 from a fuel reprocessing plant dissolver off-gas system. The off-gas has the composition of air and contains .0315% CO_2 by volume. It is flowing at 740 cfm. As in Appendix A, a packed column is to be designed for the absorption process, operating at atmospheric pressure and 20°C.

The same definitions, assumptions, parameters, and references outlined in Appendix A will be used.

System Design

The flow rate of the off-gas (air) is 740 cfm. So,

$$G' = 740 \ \frac{ft^3}{min} \times .0752 \ \frac{lb}{ft^3} \times \frac{60 \ min}{hr} = 3339 \ \frac{lb}{hr}.$$

As in the smaller column design in Appendix A, try an L' that is 81 times G'.

$$81 \times 3339 = 270{,}459 \ \frac{lb}{hr} \Rightarrow 270{,}459 \ \frac{lb}{hr} \times \frac{1 \ ft^3}{67.7 \ lb} \times \frac{7.48 \ gal}{ft^3}$$

$$\times \ \frac{1 \ hr}{60 \ min} = 498 \ gpm.$$

As before,

$$\frac{L'}{G'} \left(\frac{\rho_G}{\rho_L}\right)^{1/2} = 2.7$$

and

$$G = 229.68 \ \frac{lb}{ft^2\text{-}hr}$$

The column diameter is:

$$D = \left(\frac{3339}{229.68} \times \frac{4}{\pi}\right)^{\frac{1}{2}} = 4.3 \text{ ft.}$$

Again,

$$K_G a = 2.25.$$

The amount of CO_2 that enters the column/hr is:

$$\frac{3339\ (.000315)}{(.0752)(359)(\frac{528}{492})} = .036\ \frac{\text{lb-moles}}{\text{hr}}.$$

For 90% removal of CO_2:

$$\Delta P_{LM} = .000123.$$

$$N = .9\,(.036) = .0324\ \frac{\text{lb-moles}}{\text{hr}}$$

$$A = \frac{\pi}{4} \times (4.3)^2 = 14.52 \text{ ft}^2$$

$$h = \frac{.0324}{(2.25)(14.52)(.000123)} = 8.06 \text{ ft.}$$

The height to diameter ratio of the column is less than 2. It should be considerably larger for sound column design. This can be achieved by design alterations to the column.

Now, by cutting the cross sectional area in half and doubling the height, the same results (90% removal of CO_2) can be obtained.

So,

$$A = \frac{14.52}{2} = 7.26 \text{ ft}^2$$

$$h = 16.12 \text{ ft.}$$

Working backwards, the other parameters can be obtained.

$$D = \left(\frac{7.26 \times 4}{\pi}\right)^{\frac{1}{2}} = 3.04 \text{ ft.}$$

$$G = \frac{3339}{7.26} = 459.9 \ \frac{\text{lb}}{\text{ft}^2\text{-hr}} = .128 \ \frac{\text{lb}}{\text{ft}^2\text{-sec}}.$$

$$F(G^2) = (.128)^2 \times .68 = .0111.$$

From Figure A-1,

$$\frac{L'}{G'} \left(\frac{\rho_G}{\rho_L}\right)^{\frac{1}{2}} \cong .62 = \frac{L'}{G'} \left(\frac{.0752}{67.7}\right)^{\frac{1}{2}}.$$

$$\frac{L'}{G'} = 18.6$$

$$L' = 18.6 \times 3339 = 62{,}114.6 \ \frac{\text{lb}}{\text{hr}}.$$

or

$$62{,}114.6 \ \frac{\text{lb}}{\text{hr}} \times \frac{1 \text{ ft}^3}{67.7 \text{ lb}} \times 7.48 \ \frac{\text{gal}}{\text{ft}^3} \times \frac{1 \text{ hr}}{60 \text{ min}} = 114.38 \text{ gpm.}$$

The residence time of the liquid in the column packed with 1 inch Berl saddles ($\epsilon = .78$) is:

$$7.26 \text{ ft}^2 \times 16.28 \text{ ft} \times .78 / \left(114.38 \ \frac{\text{gal}}{\text{min}} \times \frac{1 \text{ ft}^3}{7.48 \text{ gal}}\right) = 6.03 \text{ min.}$$

Checking for flooding velocity, from Figure A-1 at flooding

$$F(G^2) = .033 = G_F^2 \times .68 \Rightarrow G_F = .22 \ \frac{\text{lb}}{\text{ft}^2\text{-sec}}.$$

So,

$$\frac{.128}{.220} = .581.$$

The column is operating at 58.1% of flooding, which is sound column design.

As can be seen in Appendix A, 99 percent removal of CO_2 can be obtained by increasing the height of the column, or increasing the diameter and liquid-gas flowrate ratio. A height of 16 feet is about the maximum desired for this application due to room size constraints at the reprocessing facility. Also, a column of 3 feet in diameter is about the largest desired for a column height of 16 feet. An alternative to adding height to the column would be to cascade additional columns in series. By adding an identical column in series with the first, 99 percent removal could be obtained. In summary, the design yields:

Size of column - 3 ft. in diameter
16.12 ft. high
Air flowrate - 740 cfm
Liquid flowrate - 114.38 gpm
Number of columns - 90% CO_2 removal - 1 column
- 99% CO_2 removal - 2 columns

APPENDIX C - REPROCESSING DISSOLVER OFF-GAS SYSTEM DESIGN (CATEGORY II)

It is desired to remove CO_2 from a fuel reprocessing plant dissolver off-gas system. The off-gas has the composition of air and contains .0315% CO_2 by volume. It is flowing at 100 cfm. As in Appendix A, a packed column is to be designed for the absorption process, operating at atmospheric pressure and 20°C.

The same definitions, assumptions, parameters, and references outlined in Appendix A will be used.

System Design

The flow rate of the off-gas (air) is 100 cfm. So,

$$G' = 100 \frac{ft^3}{min} \times .0752 \frac{lb}{ft^3} \times \frac{60 \ min}{hr} = 451 \ \frac{lb}{hr}.$$

As in the column design in Appendix A, try an L' that is 81 times G'.

$$81 \times 451 = 36{,}531 \ \frac{lb}{hr} \Rightarrow 36{,}531 \frac{lb}{hr} \times \frac{1 \ ft^3}{67.7 \ lb} \times \frac{7.48 \ gal}{ft^3}$$

$$\times \frac{1 \ hr}{60 \ min} = 67.3 \ gpm.$$

As shown in Appendix A,

$$\frac{L'}{G'} \left(\frac{\rho_G}{\rho_L}\right)^{\frac{1}{2}} = 2.7$$

and

$$G = 229.68 \ \frac{lb}{ft^2\text{-}hr} = .0638 \ \frac{lb}{ft^2\text{-}sec.}$$

The column diameter is:

$$D = \left(\frac{451}{229.68} \times \frac{4}{\pi}\right)^{\frac{1}{2}} = 1.58 \text{ ft.}$$

Again,

$K_Ga = 2.25.$

The amount of CO_2 that enters the column/hr is:

$$\frac{451\ (.000315)}{(.0752)(359)(\frac{528}{492})} = .0049\ \frac{\text{lb-moles}}{\text{hr}}.$$

For 90% removal of CO_2:

$\Delta P_{LM} = .000123.$

$$N = .9(.0049) = .0044\ \frac{\text{lb-moles } Co_2 \text{ transferred}}{\text{hr}}$$

$$A = \frac{\pi}{4} \times (1.58)^2 = 1.96 \text{ ft}^2$$

$$h = \frac{.0044}{(2.25)(1.96)(.000123)} = 8.11 \text{ ft.}$$

The residence time of the liquid in the column packed with 1 inch Berl saddles ($\epsilon = .78$) is:

$$1.96 \text{ ft}^2 \times 8.11 \text{ ft} \times .78/\left(67.3\ \frac{\text{gal}}{\text{min}} \times \frac{1 \text{ ft}^3}{7.48 \text{ gal}}\right) = 1.38 \text{ min.}$$

Checking for flooding velocity, from Figure A-1 at flooding $F(G^2)\ .0062 = G_F^2 \times .68 \Rightarrow G_F\ .095\ \frac{\text{lb}}{\text{ft}^2\text{-sec}}.$

So,

$$\frac{.0638}{.095} = .672$$

So, operation is at 67.2% of flooding, which is sound column design.

Now, if 99% removal of CO_2 is desired

$$\Delta P_{LM} = \frac{.000315 - .00000315}{\ln\left(\frac{.000315}{.00000315}\right)} = .0000677$$

$$N = .99\ (.0049) = .00485$$

$$h = \frac{.00485}{2.25\ (1.96)\ (.0000677)} = 16.24 \text{ ft.}$$

The residence time of the liquid in the column is about twice that for 90% removal since the column is about twice as tall.

The column is operating at 67.2% of flooding.

Table C-1

Desired CO^2 Removal	Liquid Flow Rate	Gas Flow Rate	Column Diameter	Column Height	Residence Time	Operational % of Flooding
90%	67.3 gpm	100 ft^3/min	1.58 ft (19 inches)	8.11 ft	1.38 min	67.2%
99%	67.3 gpm	100 ft^3/min	1.58 ft (19 inches)	16.24 ft	1.38 min	67.2%